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Strong localization of carriers in δ-doped GaAs structures

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Electrical conduction in Be δ-doped GaAs structures grown by molecular-beam epitaxy was studied in the temperature range from 10 to 350 K. A Be δ-doped layer and a 1-nm-thick GaAs spacer layer were grown at 520 °C, followed by growth of a 1-nm-thick GaAs layer with a high concentration of excess As and a 5-nm-thick nearly stoichiometric GaAs layer at a temperature close to 150 °C. At low Be doping concentrations, the conduction is *n* type and thermally activated, occurring in the 1-nm-thick low-temperature-grown GaAs layer. At higher Be doping concentrations, the conduction is *p* type and thermally activated, resulting from thermal excitation of localized holes to extended states in the δ-doped well. Activation energies for the *p*-type conduction range around 100 meV, indicating strong localization of holes in δ-doped wells. By further increasing the Be concentration, the activation energy decreases and eventually leads to the metal-like temperature dependence of the conduction at room temperature, suggesting a possible metal-insulator transition. High-temperature limits of the resistivity of insulating samples were found to be close to the value of the quantum unit of resistance, $\frac{1}{2}h/e^2$.

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I. INTRODUCTION

Metal-insulator transitions resulting from Anderson localization occur in impurity-doped semiconductors.[1] At low impurity concentrations, electrical conduction occurs via carrier hopping among impurity ions. At higher impurity concentrations, impurity bands form, and electrical conduction occurs via thermal excitation of carriers from localized states to extended states. With a further increase in the impurity concentration, the conduction becomes metallic. Excitation energies from localized states to extended states in impurity bands are typically a few meV,[2] and, hence, effects of the Anderson localization on the conduction become significant only at temperatures far below room temperature in these impurity-doped semiconductors.

Strength of the Anderson localization may be increased by a number of methods. Larger variation in random potentials results in stronger localization.[3] In lower-dimensional systems, the localization is more significant according to scaling theory.[4] A lower concentration of carriers is also expected to lead to stronger localization because of reduced screening of random potentials. During the course of investigation of electrical conduction in ultrathin GaAs layers grown at low temperatures (LT-GaAs) by molecular-beam epitaxy (MBE),[5] we have found strong localization of carriers in Be δ-doped wells, considered to result from the above three methods.

In the past, electrical transport properties of δ-doped structures have been extensively studied.[6] Among a number of combinations between impurities and host semiconductors, Si and Be δ doping in GaAs have been studied in the greatest detail.[7] According to these studies, all carriers donated by impurities contribute to electrical conduction at room temperature with their mobility increasing at lower temperatures. Thermally activated conduction resulting from carrier localization was observed in Si δ-doped GaAs only at temperatures far below room temperature with low Si concentrations,[8] similarly to the case of impurity-doped bulk semiconductors.

In the samples investigated in the present study, a Be δ-doped well is placed in the vicinity of an ultrathin LT-GaAs layer which contains a high concentration of excess As point defects. The majority of excess As point defects are antisite As atoms that form deep donor states at the middle of the band gap.[9] A high concentration of holes donated by Be atoms are, hence, trapped by antisite As atoms, resulting in a lower hole concentration in the δ-doped well. The method of removal of carriers from a δ-doped well has been widely used in selectively δ-doped $Al_xGa_{1-x}As$/GaAs heterostructures.[10] In these structures, electrical conduction in a δ-doped well is overwhelmed by electrical conduction in a GaAs quantum well and, hence, cannot be experimentally observed. In LT-GaAs, on the other hand, electrical conduction occurs via hopping of electrons among antisite As atoms.[11] At room temperature, mobility of hopping electrons in LT-GaAs is very low, around 0.1 cm^2/V s,[12] and its bulk resistivity is higher than 10 Ω cm.[11] With the samples used in the present study, therefore, it has become possible to reveal electrical transport properties of carriers remaining at deep levels of a δ-doped potential well.

From our samples, thermally activated conduction with activation energies of nearly 100 meV has been observed at room temperature, indicating strong localization of holes in the δ-doped well. To our best knowledge, such strong localization of carriers has not been reported in the past for impurity-doped semiconductors. The strong localization of holes in the δ-doped well is attributed to a large variation of a random potential caused by negatively charged Be ions and positively charged antisite As atoms as well as by reduced screening of the random potential by a low concentration of holes. By increasing the Be doping concentration to 10^{14} cm^{-2}, the activation energy gradually decreases towards zero with nearly constant high-temperature limits of the resistivity, suggesting a possible metal-insulator transi-

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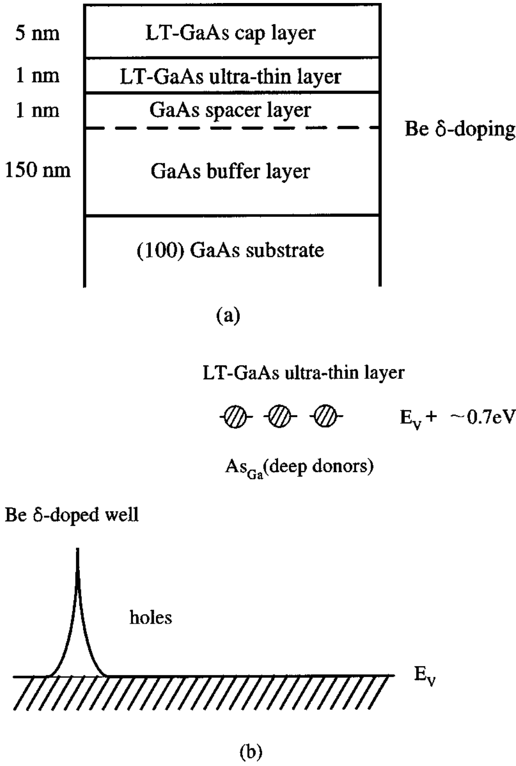

FIG. 1. (a) Structure of the δ-doped GaAs samples, and (b) energy diagram of a Be δ-doped well and antisite As atoms, As_{Ga}. Layer thicknesses are shown on the left-hand side of (a). Shaded regions represent the states occupied by electrons.

tion. As an important observation, high-temperature limits of the resistivity of insulating samples were found to be close to the quantum unit of resistance, $\frac{1}{2}h/e^2$. In this paper we present these experimental results, which have been obtained by van der Pauw resistivity and Hall-effect measurements of MBE-grown δ-doped structures.

II. EXPERIMENT

Figure 1 schematically shows a sample structure and an energy diagram corresponding to this structure without transfer of carriers. The Be δ doping and growth of a GaAs spacer layer were carried out at 520 °C, and, therefore, doped Be atoms are expected to act as acceptors[6] and form a narrow triangularlike potential well for holes as shown in Fig. 1(b). According to theoretical calculations,[6] the energy of the lowest state in a δ-doped well is proportional to $N^{2/3}/m^{*1/3}$, where N and m^* are the dopant concentration and effective mass, respectively. For Be concentrations used in the present study, these energies are a few hundred meV. In the ultrathin LT-GaAs layer antisite As atoms, As_{Ga}, form midgap donor states whose energy levels are known to be located at approximately $E_v+0.7$ eV where E_v is the valence-band edge.[9]

Samples were grown by utilizing a conventional MBE system. Semi-insulating epiready (100) GaAs wafers were used as substrates and mounted on a Mo holder with indium. A Ga flux used throughout growth experiments was 5.8×10^{-7} Torr, which gave rise to a growth rate of 0.9 μm/h. After desorption of an oxide layer of the substrate surface, the surface was annealed for 10 min at 600 °C, followed by growth of a 150-nm-thick GaAs buffer layer at 580 °C. After growth of the buffer layer, the sample temperature was lowered to 520 °C for Be δ doping and growth of a GaAs spacer layer with a thickness of 1 nm. The lower sample temperature was chosen for minimizing diffusion and surface segregation of Be,[6] but as explained later the diffusion and surface segregation occurred to a certain extent for very high doping concentrations.

Immediately after growth of the spacer layer, the sample temperature was lowered for growth of LT-GaAs layers. For setting the growth temperature, the sample temperature was first set at 200 °C by using a pyrometer[13] and then further lowered by 50° confirmed by reading a thermocouple placed behind the substrate holder. The thermocouple reading of the sample temperature set in this way ranged from 135 °C to 145 °C. An ultrathin LT-GaAs layer with a thickness of 1 nm was grown with an As flux of 3.0×10^{-5} Torr followed by growth of a LT-GaAs cap layer with a thickness of 5 nm. The condition used for the growth of the ultrathin LT-GaAs layer is known to give rise to a high concentration of antisite As atoms without forming the extended defects of our earlier study.[5] The concentration of antisite As atoms in ultrathin LT-GaAs layers grown under similar conditions was found to range from 2×10^{20} to 4×10^{20} cm^{-3} by x-ray-diffraction analyses.[5,14] An As flux for growth of the GaAs cap layer is 6.2×10^{-6} Torr, which is known to give rise to a nearly stoichiometric LT-GaAs layer with the aforementioned growth rate.[13] The cap layer was grown in order to prevent oxidation of the ultrathin LT-GaAs layer in air. Reflection high-energy electron-diffraction (RHEED) patterns observed after growth of cap layers showed that the layers were singly crystalline without any indication of formation of extended defects. Beryllium concentrations in the samples were estimated with Be effusion cell temperatures and deposition times of Be on GaAs surfaces. Beryllium flux intensities for given effusion cell temperatures were calibrated by growing uniformly doped layers at 580 °C and measuring their hole concentrations.

A square 5×5-mm sample was cut for the van der Pauw and Hall-effect measurements, and an In contact was made at each corner of a sample. No annealing was made for making contacts, but good ohmic contacts were obtained in all samples. Resistivity measurements were made with samples remaining on substrates whose thicknesses were 0.6 mm. The contribution of electrical conduction of the substrate to the measured resistivity of the samples was, however, negligible because of the very high resistivity of the substrate. The sheet resistance of the substrate with a 150-nm-thick buffer layer on it was 8.0×10^7 Ω at room temperature, while sheet resistance of all measured samples in this study was less than 5×10^6 Ω at room temperature. No correction, therefore, was made for the contribution of the substrate and buffer layer to the measured resistivity.

III. RESULTS AND DISCUSSION

Electrical conduction of a number of samples, which had no δ-doped layer but only ultrathin LT-GaAs layers and LT-

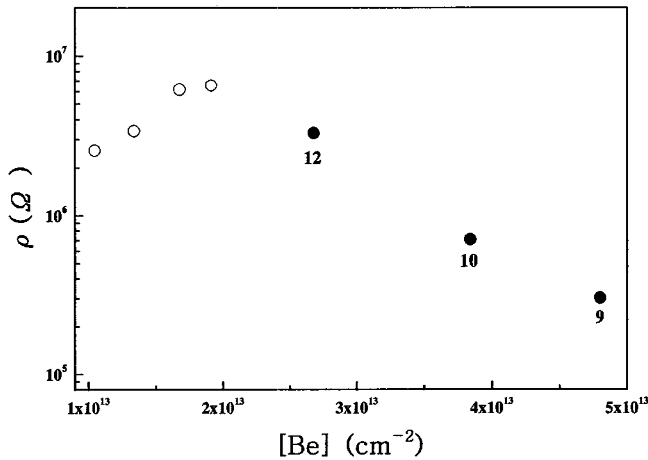

FIG. 2. Room-temperature resistivity of δ-doped samples as a function of the Be doping concentration. Open circles indicate the n-type conduction, while solid circles represent the p-type conduction. Numbers attached to solid circles correspond to sample numbers listed in Table I.

GaAs cap layers, was first examined. All these nondoped samples exhibit thermally activated n-type conduction, known to occur by hopping of electrons among antisite As atoms.[11] Their resistivity at room temperature as two-dimensional systems ranges from 1.0×10^6 to 2.0×10^6 Ω, and activation energies for the conduction are close to 100 meV.

Introduction of Be δ doping with concentrations higher than 1.0×10^{13} cm^{-2} results in significant changes in the electrical conduction. Figure 2 shows room-temperature resistivity of a number of doped samples as a function of the Be doping concentration. In the figure, open circles indicate the n-type conduction, while solid circles represent the p-type conduction. As seen in the figure, the resistivity initially increases with the Be concentration but starts to decrease rapidly once the conduction type changes into the p type. The increase in the resistivity in the n-type region is ascribed to the reduction of electrons for hopping conduction among antisite As atoms in the ultrathin LT-GaAs layer due to transfer of electrons to the Be δ-doped well. This implies that in this n-type region the Be δ-doped well is depleted of holes, or a hole concentration in the well is too low to give rise to any significant conduction. The reduction of the hopping-electron concentration with an increase of Be doping may be directly observed by Hall-effect measurements of these n-type samples, but accurate values of electron concentrations could not be estimated by this method because of very low mobility of hopping electrons. The increase in the resistivity towards the transition from the n- to the p-type conduction, however, strongly suggests that the above-mentioned transfer of carriers and resulting compensation occur in the ultrathin LT-GaAs and δ-doped layers. To show another supporting result, it is noted that concentrations of antisite As atoms in ultrathin layers range from 2×10^{20} to 4×10^{20} cm^{-3}, and, hence, their sheet concentrations are close to the Be concentration where the conduction type changes from n to p. The p-type conduction at a higher Be concentration, therefore, can be attributed to holes that remain in the δ-doped well after accommodating electrons from antisite As atoms. The p-type conduction at room temperature in all samples included in Fig. 2 is thermally activated with activation energies higher than several tens of meV.

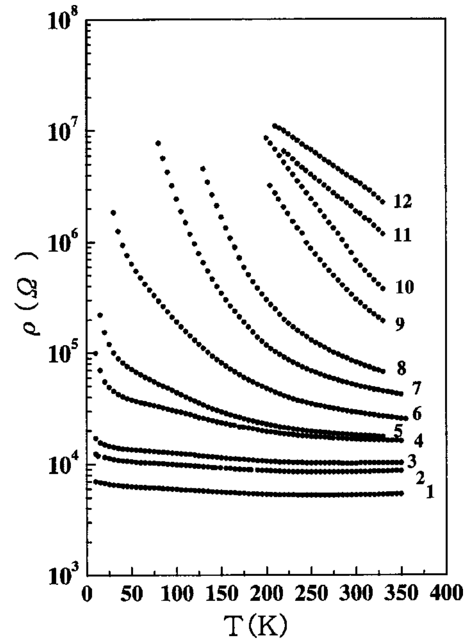

FIG. 3. Temperature dependence of the resistivity of 12 p-type samples.

Figure 3 shows temperature dependence of the resistivity of 12 p-type samples. In Table I, Be doping concentrations and room-temperature resistivity, Hall mobility, and activation energies of the samples are listed. As seen in Table I, the resistivity monotonically decreases with increases in the Be concentration. There are, however, some opposite cases where the resistivity slightly increases in spite of an increase in the Be concentration. This inconsistency is believed to be caused by the difficulty of the accurate control of concentrations of antisite As atoms in the ultrathin LT-GaAs layer. The concentration of antisite As atoms is known to vary with a slight change in the growth temperature of the ultrathin LT-GaAs layer,[5] the accurate control of which is difficult under the present experimental conditions.

There is another problem that needs to be pointed out with respect to Be concentrations in Table I. Beryllium atoms are known to diffuse rapidly in GaAs, and, hence, δ doping of Be at a high temperature around 600 °C results in significant broadening of the concentration profile.[6] In order to minimize such diffusion, the δ doping and the growth of the spacer layer were carried out at 520 °C. Even at this lower temperature, significant diffusion and surface segregation were found to occur at very high doping concentrations. After Be δ doping with concentrations higher than the middle of 10^{13} cm^{-2}, threefold superstructure reflections appeared in the $[0\bar{1}1]$ RHEED patterns, which were known to result from surface reconstruction induced by Be deposition.[15] During growth of the GaAs spacer layer after δ doping, these threefold reflections disappeared but reappeared soon after

TABLE I. Beryllium doping concentration and room-temperature (RT) resistivity, Hall mobility, and activation energy of 12 samples.

Sample	[Be] (cm^{-2})	ρ_{RT} (Ω)	μ_{RT} (cm^2/V s)	ε_{RT} (meV)
1	1.54×10^{14}	5.22×10^{3}	53.9	
2	1.13×10^{14}	8.47×10^{3}	43.3	
3	1.25×10^{14}	1.01×10^{4}	56.1	
4	9.60×10^{13}	1.57×10^{4}	74.1	7.2
5	1.06×10^{14}	1.81×10^{4}	57.4	9.8
6	8.64×10^{13}	2.83×10^{4}	67.5	23
7	6.72×10^{13}	4.97×10^{4}	89.4	34
8	5.76×10^{13}	8.22×10^{4}	97.7	60
9	4.80×10^{13}	3.03×10^{5}	107	130
10	3.84×10^{13}	7.08×10^{5}	80.7	150
11	1.81×10^{13}	1.93×10^{6}	8.99	92
12	2.68×10^{13}	3.29×10^{6}	4.98	83

the growth of the spacer layer. This implies that significant diffusion of Be through the spacer layer to the surface or severe surface segregation of Be have occurred during growth of the spacer layer. The accurate value of the Be concentration in the δ-doped well of each sample, therefore, is not known. The concentrations listed in Table I are only roughly estimated values of the Be concentrations in δ-doped wells, but the actual Be concentrations in δ-doped wells are expected to increase with the increase of these listed concentrations. The uncertainty of absolute values of Be concentrations in δ-doped wells, hence, will not affect main conclusions derived in the present paper.

There are two possible explanations for the origin of the thermally activated *p*-type conduction. The first explanation assumes that a Be δ-doped well is completely depleted of holes. The *p*-type conduction occurs as a result of thermal excitation of holes from midgap states of antisite As atoms in the ultrathin LT-GaAs layer to the lowest state in the δ-doped well. The activation energy corresponds to the energy difference between the midgap states and the lowest state in the δ-doped well. The second explanation assumes localization of holes at deep levels in the δ-doped well. The *p*-type conduction occurs via thermal excitation of holes from localized states to extended states in the δ-doped well.

In the first case the activation energy for the conduction is expected to decrease monotonically with increase in the Be doping concentration. As explained earlier, the energy of the lowest state in the δ-doped well is proportional to $N^{2/3}$. Hence, the energy difference between the lowest state and the midgap states of antisite As atoms decreases as the δ-doped well becomes deeper. Figure 3, however, shows a different trend from this prediction. Activation energies of samples 11 and 12 are significantly lower than those of samples 9 and 10 as seen in Table I. All other samples that have similar Be concentrations to those of samples 11 and 12 but are not included in Fig. 3 also exhibit low activation energies. The activation energy of the *p*-type conduction, therefore, is initially low for a sample with a low Be concentration but increases to a high value, then gradually decreases to a lower value with a further increase in the Be concentration.

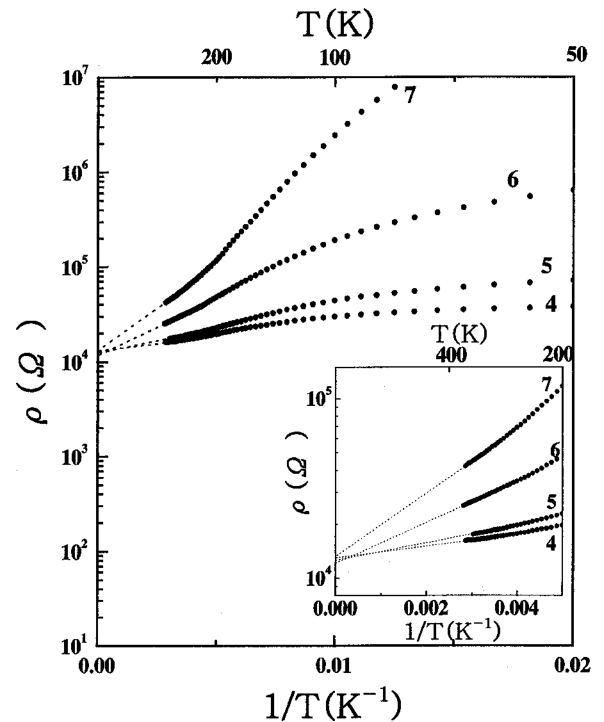

FIG. 4. Resistivity of samples 4–7 as a function of $1/T$.

From earlier studies of the impurity conduction in doped semiconductors,[1] it is known that at lower impurity concentrations the conduction occurs via carrier hopping among localized states, and at higher impurity concentrations the hopping conduction is replaced by the conduction via thermal excitation from localized states to extended states. At similar impurity concentrations, activation energies of the former type of conduction are lower than those of the latter type of conduction. Because of much higher mobility of carriers in extended states than that of the hopping conduction, the conduction via the excitation to extended states overwhelms the hopping conduction in spite of the higher activation energy at a certain impurity concentration. In agreement with this physical picture, Table I shows significantly low Hall mobility of samples 11 and 12 compared to that of samples 9 and 10. In samples 11 and 12, a low concentration of holes is expected to remain in the δ-doped well as a result of transfer of the majority of holes to the ultrathin LT-GaAs layer and are likely to stay at deep localized states. It may, therefore, be concluded that the thermally activated *p*-type conduction occurs in the δ-doped well via hopping of holes among localized states at low Be concentrations and via thermal excitation to extended states at high Be concentrations.

Figure 4 plots logarithms of resistivity of samples 4–7 as a function of the reciprocal of the temperature. According to the above explanation, in these four samples the conduction occurs via thermal excitation of holes from localized states to extended states in the δ-doped well. As seen in Fig. 4, the slope of each curve is not constant in the entire temperature range. For samples 4–6, slopes in the low-temperature side are smaller than those in the high-temperature side. This sug-

gests that, as in the case of doped bulk semiconductors, the conduction in the low-temperature side may be due to hopping of carriers instead of excitation to extended states. In such a case, the logarithms of the resistivity may change linearly with $T^{-1/3}$, as expected from variable range hopping in two dimensions.[16] We, however, will not discuss this possibility further in this paper.

The logarithms of resistivity of all four samples in the temperature range above 250 K exhibit nearly linear changes with $1/T$ as seen in the inset of Fig. 4. In addition, the figure shows that extrapolated straight lines from the data in this temperature range appear to converge onto a single value at $1/T=0$. If the curves in the high-temperature range do become linear, the results are consistent with infinite-temperature intercepts, namely, high-temperature limits of resistivity, close to $\frac{1}{2}h/e^2$, which is 12 906 Ω. The extrapolated values derived by least-square fits for the data of the four samples above 250 K range from 11 863 to 12 774 Ω. The standard deviation of the high-temperature limit of each sample that results from the least-square fit is less than 1%. For samples 8–10, measured temperature ranges where logarithms of resistivity change linearly with $1/T$ are too narrow to give rise to accurate values of high-temperature limits of resistivity.

Nearly a single value of high-temperature limits of the resistivity of the above four samples implies that the preexponential factor of the Arrhenius-type expression of the resistivity remains constant, while the activation energy gradually decreases towards zero with Be concentration. This suggests that, at the hole concentration where the activation energy vanishes, a transition occurs from a phase in which carriers are strongly localized to a phase without such strong localization. The room-temperature resistivity of samples 1–3 is lower than the high-temperature limits of the resistivity of the above four samples. The resistivity of these three samples exhibits a gradual increase at lower temperatures in Fig. 3, suggesting that they are insulating samples, like other samples. Close inspection of the temperature dependence of the resistivity of these samples, however, revealed that it was metal-like in the high-temperature range. Figure 5 shows the temperature dependence of the resistivity of samples 1–3 in the high-temperature range. The resistivity of samples 1 and 2 increases with temperature in the temperature range above 260 K, below which the resistivity exhibits the opposite temperature dependence. The resistivity of sample 1 at 260 K is 5.24 kΩ, while its resistivity at 350 K is 5.36 kΩ. The resistivities of sample 2 at 260 and 350 K are 8.50 and 8.67 kΩ, respectively. The resistivity of sample 3, which is closer to the above high-temperature limits, remains nearly constant in this temperature range as seen in Fig. 5. With this metal-like temperature dependence of the resistivity, it may be suggested that a possible metal-insulator transition occurs at room temperature with the change in the hole concentration in this system. It should, however, be pointed out that an increase in the resistivity with the temperature also occurs as a result of carrier-phonon scattering, and, therefore, further experimental studies are necessary in order to clarify if samples 1–3 are in a metallic phase or a phase with weaker localization at room temperature.

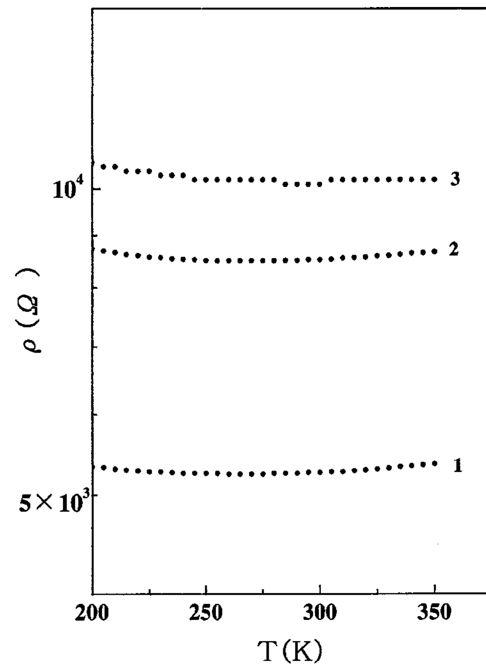

FIG. 5. Temperature dependence of the resistivity of samples 1–3 in the high-temperature range.

As stated in the Introduction, Ye *et al.* have observed thermally activated conduction in Si δ-doped GaAs samples,[8] indicating carrier localization in their samples. The carrier localization they observed, however, is significantly different from the present case. In their samples, Si was δ doped in GaAs layers under the normal MBE growth condition, so that equal numbers of conduction electrons and Si ions existed in a δ-doped well, resulting in rather small potential variations in the well. The thermally activated conduction, therefore, occurred in the samples with very low doping concentrations at low temperatures. In the present case, on the other hand, a high concentration of deep donor antisite As atoms is present in the vicinity of a Be δ-doped well. Hence, a significant portion of holes donated by δ-doped Be are trapped by antisite As atoms. The resulting state may be described as follows. High concentrations of negatively charged Be ions and positively charged excess As point defects form a pair of sheets with a spacing of only 1 nm. A low concentration of holes occupy deep states in a narrow potential well caused by Be ions. Because of large potential variations caused by random arrangements of Be ions and ionized antisite As atoms, holes are strongly localized in the δ-doped well. Extended states that contribute to the conduction are also considered to be located at relatively deep levels in the δ-doped well, so that the system may be considered two dimensional even at room temperature as explained below.

The high-temperature limits of the resistivity of the samples with low activation energies are close to the value of $\frac{1}{2}h/e^2$. The samples whose resistivities are lower than this value also exhibit metal-like temperature dependence of the resistivity in the high-temperature range. In a number of two-dimensional systems where metal-insulator transitions were found to occur in recent years, the resistivity at the separatrix

between metallic and insulating behavior is of an order of magnitude of h/e^2.[17] In two-dimensional systems where thermally activated conduction occurs, high-temperature limits of the resistivity were found to be close to the quantum unit of resistance.[18,19] In particular, the high-temperature limit of the resistivity in the variable range hopping regime of a Si δ-doped GaAs/$Al_xGa_{1-x}As$ heterostructure is nearly equal to $\frac{1}{2}h/e^2$,[19] such as in the present case. The closeness of the high-temperature limits of the resistivity to the quantum unit of resistance in the present case, therefore, seems to result from the fundamental nature of two-dimensional electron systems. It should, however, be pointed out that the resistivity was measured in the temperature range below the liquid-helium temperature in all aforementioned reported studies, while it was measured at by far higher temperatures in the present case. The similarity between the present case and other reported cases, hence, may be surprising and needs to be investigated closely.

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