Anomalous Hall effect of metallic Be/Si pair δ-doped GaAs structures

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Metallic samples of Be and Si pair δ-doped GaAs structures which undergo a metal-insulator transition with a decrease in the hole concentration are investigated by Hall resistance and magnetoresistance measurements. The anomalous Hall effect and negative magnetoresistance are observed from the samples in a temperature range above 70 K. Magnitudes of negative magnetoresistance and anomalous Hall resistance significantly vary among the samples, although their doping conditions are close to one another. Dependence of anomalous Hall resistance on the temperature and applied magnetic field is closely correlated to that of negative magnetoresistance for each sample. Their dependence is explained on the basis of a paramagnetic state of localized magnetic moments coexisting with itinerant holes in these samples. Both anomalous Hall effect and negative magnetoresistance decrease with lowering the temperature from 150 K and vanish at a temperature around 70 K, a possible origin of which is discussed.

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I. INTRODUCTION

In the past decade, a number of spin-related phenomena have been actively investigated in the field of semiconductor physics, partly driven by a new approach called spintronics. Their earlier representative is ferromagnetism of diluted magnetic semiconductors based on III-V compounds.[1] More recent studies on such phenomena include those on the spin Hall effect,[2] spontaneous spin polarization in quantum point contacts,[3] and enhanced Pauli spin susceptibility of two-dimensional electron systems.[4] These studies suggest that electron systems in semiconductors are capable of giving rise to a variety of novel spin-related phenomena, particularly in their highly controlled low-dimensional structures.

Semiconductors doped with shallow-level impurities such as P-doped Si have been known to give rise to spin-related phenomena since the early time of the field of semiconductor physics. Electron-paramagnetic-resonance studies provided direct evidence of the existence of localized magnetic moments associated with hydrogenic impurity states.[5] By an electron-nuclear-double-resonance study, these magnetic moments were found to undergo antiferromagnetic coupling with each other.[6] Localized magnetic moments in impurity-doped semiconductors have been further studied in connection with a metal-insulator transition which occurs with an increase in the impurity concentration.[7] According to these studies, the temperature dependence of magnetic susceptibility of insulating samples which are away from a metal-insulator transition is essentially ascribed to localized magnetic moments associated with isolated hydrogenic impurity states. It has been, however, found that the existence of localized magnetic moments persists to metallic samples where they may no longer be considered simply as those associated with isolated hydrogenic impurity states.[8] Theoretical studies have indicated that the existence of such localized magnetic moments results from the intrinsic nature of a disordered metallic phase in the vicinity of a metal-insulator transition.[9] These localized magnetic moments can be observed only at low temperatures because of thermal excitation of carriers from shallow impurity levels to either conduction or valence bands.

In order to explore new possibility of localized magnetic moments induced by doping of nonmagnetic impurities in semiconductors, we have recently carried out magnetoresistance measurements of Be δ-doped GaAs structures grown by molecular-beam epitaxy (MBE).[10] The structure is made of a pair of a Be δ-doped layer and a donor δ-doped or ultrathin layer which has a higher Be concentration than a donor concentration. Beryllium atoms and donor atoms compensate each other, leaving remaining holes at deep levels in the Be δ-doped potential well. These holes are strongly localized, resulting in thermally activated conduction at room temperature.[11,12] The strong localization of holes is ascribed to a narrow potential well for deep levels of the δ-doped layer and a severe random potential caused by high concentrations of negatively charged Be ions and positively charged donor ions; a hydrogenic impurity state is squeezed into a quasi-two-dimensional form in the narrow potential well and hence have a larger binding energy of a hole.[13] With an increase in the Be concentration with respect to the donor concentration, the activation energy for the conduction decreases and eventually results in a metal-insulator transition of the quasi-two-dimensional system.[11] The change of the activation energy from highly insulating samples to those near the metal-insulator transition, which corresponds to the change of the Fermi level with respect to the mobility edge, ranges over a few tens of meV.[11] The pair δ-doped structure, hence, is considered to exhibit carrier-localization phenomena similar to those of impurity-doped bulk semiconductors but have a larger energy scale than that of the latter system. It is, therefore, expected that localized magnetic moments may exist in the pair δ-doped structure at higher temperatures than those where they are observed in impurity-doped bulk semiconductors.

In the aforementioned magnetoresistance study,[10] insulating samples were mainly investigated. Negative magnetoresistance was observed from all investigated samples in the temperature range from approximately 100 K to room temperature with an applied magnetic field parallel to δ-doped layers. On the basis of the similarity of this system to impurity-doped bulk semiconductors, the observed negative magnetoresistance was explained as a result of reduced scat-

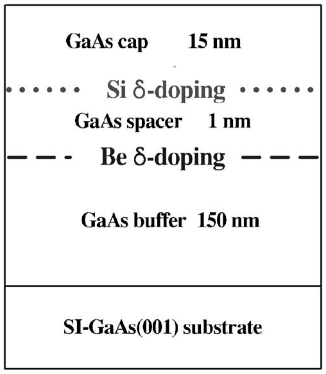

FIG. 1. Layer configuration of the Be and Si pair δ-doped structure with layer thicknesses.

tering of carriers by localized magnetic moments associated with hydrogenic impurity states at Be atoms.

In this paper, we present results of a magnetotransport study which has been focused on metallic samples of a Be and Si pair δ-doped structure. In a metallic sample, itinerant holes exist. If localized magnetic moment exists in a metallic sample, stronger interaction between carriers and localized magnetic moments is expected to occur in comparison to the case of insulating samples; in the latter samples, carriers are made by thermal excitation of localized holes to high-energy extended states.

Negative magnetoresistance indicating the existence of localized magnetic moments was observed from these metallic samples. The samples which showed large negative magnetoresistance gave rise to anomalous Hall resistance, the observation of which is attributed to stronger interaction between carriers and localized magnetic moments in these samples. The dependence of the anomalous Hall resistance on the magnetic field and the temperature is closely correlated to that of the negative magnetoresistance and is explained on the basis of a paramagnetic state of localized magnetic moments. These results which were obtained from two different kinds of magnetotransport measurements have strongly suggested the existence of localized magnetic moments in the pair δ-doped structure at high temperatures.

II. EXPERIMENT

As donor species of pair δ-doped GaAs structures, we have used Se, Si, and antisite As in the past studies.[11,12] Among these donor species, Si has the highest controllability of its doping concentration in the MBE growth. In this study, we have grown a number of samples of the Be/Si pair δ-doped structure under closely varied doping conditions. In Fig. 1, the sample structure is schematically shown. Two δ-doped layers, Be and Si, a 1-nm-thick spacer layer, and a cap layer were grown at 400 °C. Detailed growth conditions were reported in an earlier paper.[12] Results obtained from six samples listed in Table I are presented in this paper. All samples exhibit the p-type conduction. Among the samples, sample 1 has only a single Be δ-doped layer. Electronic properties of a single Be δ-doped layer were extensively studied in the past.[14] This sample was hence included as a reference sample in which almost all holes are known to be in extended states for the Be concentration used here. The doping concentrations of Be and Si were estimated from shutter opening times of δ doping with carrier concentrations in thick-layer samples where Be and Si were doped uniformly with the same cell temperatures.

TABLE I. Beryllium and Si δ-doping concentrations and room-temperature sheet resistance of six samples.

Sample	[Be] (cm^{-2})	[Si] (cm^{-2})	R_s $(\Omega)_{(300\ K)}$
1	4.90×10^{13}		3.230×10^{3}
2	4.15×10^{13}	1.01×10^{13}	5.928×10^{3}
3	4.15×10^{13}	0.90×10^{13}	6.257×10^{3}
4	4.15×10^{13}	1.23×10^{13}	6.868×10^{3}
5	3.32×10^{13}	0.67×10^{13}	7.611×10^{3}
6	2.90×10^{13}	0.67×10^{13}	9.973×10^{3}

A square 5×5 mm^2 sample was cut for the van der Pauw measurements of sheet resistance and Hall-effect measurements, and In contact was made at each corner of a sample.[11] For magnetoresistance and Hall resistance measurements, a rectangular 3×8 mm^2 sample was cut, and four In contacts with an equal spacing in the longitudinal direction and two In contacts in the transverse direction were made for the former and latter measurements, respectively.

III. RESULTS

Figure 2 shows the temperature dependence of sheet resistance R_s of six samples listed in Table I. The sheet resistance R_s was measured by using van der Pauw samples. As reported in our earlier papers, the room-temperature sheet resistance which divides insulating and metallic samples of the pair δ-doped GaAs structures was found to be close to the quantum unit of resistance $\frac{1}{2}h/e^2$, 12.9 kΩ.[11,12] All five

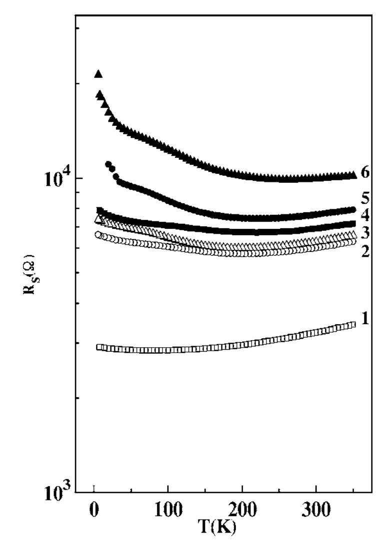

FIG. 2. Temperature dependence of sheet resistance R_s of six samples.

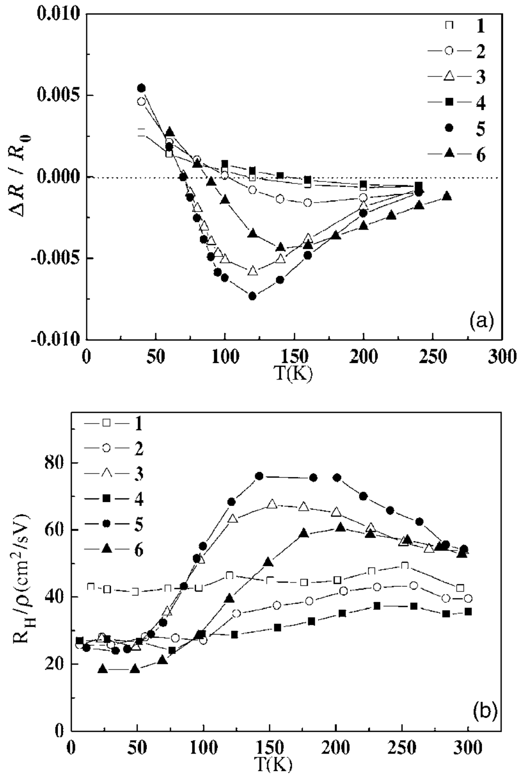

FIG. 3. (a) Temperature dependence of normalized magnetoresistance $\Delta R/R_0$, where ΔR is the change in the longitudinal resistance from the zero-field resistance R_0 by the application of the magnetic field of 9 T. (b) Temperature dependence of the Hall coefficient R_H divided by the resistivity ρ. The magnetic field of 0.32 T was applied for the Hall-effect measurements.

samples of the pair δ-doped structure have room-temperature sheet resistance lower than this value and hence correspond to metallic samples. As explained later, the Hall mobility of these samples remains higher than 20 cm^2/s V down to 5 K, further indicating that they are metallic samples. In the case of insulating samples, the mobility becomes lower than 1 cm^2/s V at such low temperatures, resulting from the dominance of hopping conduction due to complete localization of carriers.[11] Sample 1 shows the positive temperature dependence of the sheet resistance in the whole temperature range, indicating that almost all holes are in extended states in this sample.

Figure 3(a) shows the temperature dependence of ratios $\Delta R/R_0$ of six samples, where ΔR is a change in the longitudinal resistance from the zero-field resistance R_0 by application of a magnetic field of 9 T. The magnetic field B was applied in the direction parallel to the current and hence to the δ-doped layers. For obtaining these data, the resistance was measured as a function of the magnetic field in the range of −9–9 T at each temperature. All six samples exhibit negative magnetoresistance in the high-temperature range, and their magnetoresistance changes into positive values at lower temperatures. Magnitudes of the negative magnetoresistance are significantly different among samples. Samples 3 and 5 exhibit large negative magnetoresistance, while the sample with a single Be δ-doping layer and samples 2 and 4 show almost negligible magnitudes of negative magnetoresistance.

Figure 3(b) presents the results of Hall-effect measurements of six samples. For the measurements, van der Pauw samples were used with an applied magnetic field of 0.32 T. The figure plots the Hall coefficient R_H divided by resistivity ρ as a function of the temperature. If only the normal Hall effect occurs, R_H/ρ corresponds to the Hall mobility. As seen in the figure, values of R_H/ρ of samples 2–6 are significantly different among samples in the high-temperature range, but they converge to approximately 25 cm^2/s V at temperatures lower than 70 K. It is seen that there is close correspondence between Figs. 3(a) and 3(b) in their high-temperature ranges. A shape of negative magnetoresistance curve of each sample is very similar to that of R_H/ρ, if the sign of the former is reversed. Relative magnitudes of negative magnetoresistance of five samples (samples 2–6) closely correspond to those of R_H/ρ, if approximately 25 cm^2/s V is subtracted from the latter. It should be pointed out that the aforementioned similarity was obtained from two sets of measurements which used significantly different magnitudes of applied magnetic fields, namely, 9 and 0.32 T.

There is further correspondence between results of the magnetoresistance and Hall resistance measurements. Figure 4 shows plots of Hall resistance R_{Hall} of samples 3 and 5 as function of the magnetic field for different temperatures. Hall resistance curves measured at 180, 140, and 100 K deviate from the linear relation at high magnetic fields. The curves measured at 50 K, on the other hand, are completely straight. These Hall resistance curves were obtained by calculating $[R_{Hall}(B)-R_{Hall}(-B)]/2$ with experimental values of $R_{Hall}(B)$ and $R_{Hall}(-B)$ for each magnetic field B in order to remove contributions of the magnetoresistance to the Hall resistance.

Figure 5 shows magnetoresistance curves of the same samples for different temperatures. The magnetic field was applied in the direction parallel to the current and δ-doped layers. The magnetoresistance curves measured at 140, 100, and 80 K exhibit deviation from the parabolic curves at high magnetic fields. The curve measured at 60 K, on the other hand, shows positive magnetoresistance and is completely parabolic. The deviation of magnetoresistance curves from parabolic curves occurs in the same temperature range with that where Hall resistance curves deviate from the linear relation. This temperature range also corresponds to that where these two samples exhibit large values of $-\Delta R/R_0$ and R_H/ρ in Figs. 3(a) and 3(b), respectively.

IV. DISCUSSION

Close correlation among the results shown in Figs. 3(a), 3(b), 4, and 5 suggests that there is an intrinsic property of these samples which underlies all the magnetotransport processes related to these results. In the following, we show that the close correlation of the experimental results can be reasonably explained on the basis of interacting localized magnetic moments in a paramagnetic state. Hall resistivity of a sample with localized moments in a paramagnetic state is given by the normal term and anomalous term:[15]

$$\rho_{Hall} = R_H^0 B + c\rho^n \mu_0 M, \quad (1)$$

where R_H^0 is the normal Hall coefficient, ρ resistivity, M magnetization, μ_0 permeability of free space, and c the con-

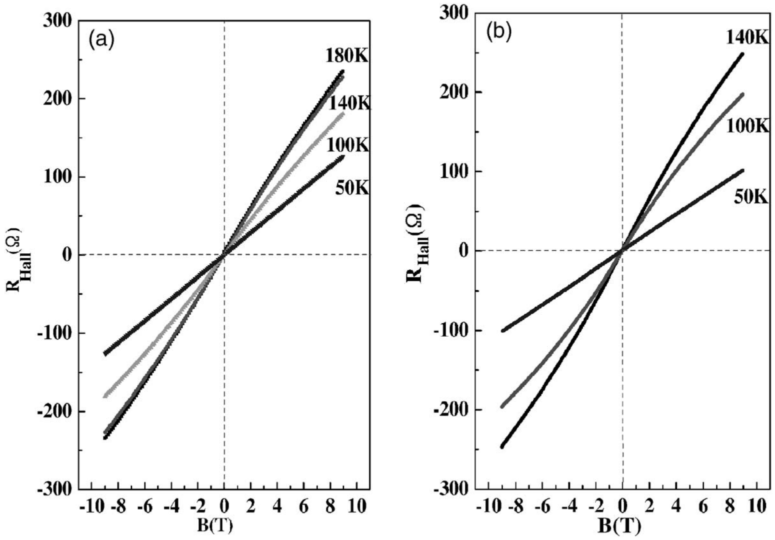

FIG. 4. Hall resistance R_{Hall} of (a) sample 3 and (b) sample 5 as a function of the magnetic field for 180, 140, 100, and 50 K.

stant. The value of the index n depends on the scattering mechanism responsible for the anomalous Hall effect; $n=1$ in the case of the skew scattering mechanism which is known to become dominant in high-mobility materials such as semiconductors.[15] The magnetization M is given by

$$M = Ng\mu_B\langle S\rangle, \tag{2}$$

where N is the concentration of localized magnetic moments, μ_B the Bohr magneton, g the Landé g factor, and $\langle S\rangle$ the thermal average of localized spins. The spin-disorder-scattering resistivity ρ_s due to localized magnetic moments, on the other hand, is given by[16]

$$\rho_s = 2\pi^2\frac{k_F m^2 J_{ex}^2}{pe^2h^3}Ng^2\mu_B^2[S(S+1) - \langle S\rangle^2], \tag{3}$$

where k_F is the Fermi wave number, m the effective mass of carrier, J_{ex} the exchange integral between a carrier spin and localized spin, p the carrier concentration, and S the localized spin.

From Eqs. (1)–(3), one can see that the anomalous term of R_H/ρ is proportional to $N\langle S\rangle$. The ratio $\Delta R/R_0$, on the other hand, is proportional to $N\langle S\rangle^2$, if the zero-field resistance R_0 is assumed to be caused only by the spin-disorder scattering. There are other significant scattering processes such as phonon and impurity scattering in these samples. Their scattering processes other than spin-disorder scattering, however, are considered to be similar among five samples of the pair δ-doped structure, as seen from nearly identical values of R_H/ρ in the low-temperature range of Fig. 3(b). As explained below, only the normal Hall effect occurs in this low-temperature range for these samples, and hence R_H/ρ in this low-temperature range corresponds to the Hall mobility. This implies that the difference of R_0 among these samples results predominantly from the difference in the carrier concentration, that is, p in Eq. (3). We can, therefore, consider that the ratio $\Delta R/R_0$ is approximately proportional to $N\langle S\rangle^2$ for these samples. On the basis of these proportional relations, one can therefore explain that relative magnitudes of $\Delta R/R_0$ of the six samples in Fig. 3(a) closely correspond to those of R_H/ρ in Fig. 3(b), where these samples have different values of N.

Both negative magnetoresistance and anomalous Hall resistance decrease and vanish at lower temperatures, as seen in Figs. 3(a) and 3(b). The most likely reason for these results is freezing of localized magnetic moments due to their direct interaction at low temperatures. Localized magnetic moments associated with hydrogenic impurity states in semiconductors are known to perform direct antiferromagnetic exchange interaction with each other.[6] At low temperatures where nearly all localized magnetic moments are considered to freeze, R_H/ρ is considered to correspond solely to the normal term, that is, the Hall mobility.

In the molecular-field approximation,[17] the magnetization M of an antiferromagnetic crystal in the magnetic field B at a temperature above the Néel temperature is given by

$$M = Ng\mu_B S B_s\left[\frac{g\mu_B S(-AM + B)}{kT}\right], \tag{4}$$

where $B_s(x)$ is the Brillouin function and

$$A = \frac{2}{g^2\mu_B^2 N}\sum_m J(R_m). \tag{5}$$

Here, $J(R_m)$ is the exchange integral and the summation is taken over nearest-neighbor spins located at R_m with respect to the reference spin. In the present case, localized spins are randomly distributed, and hence the magnitude of J_m varies continuously over a certain range. As the temperature is lowered, pairs of localized spins with larger J_m initially freeze by forming antiferromagnetic coupling, leaving others with smaller J_m as free spins. In Eq. (4), N represents the concentration of free spins and hence varies as a function of the temperature in the present case. By expanding the Brillouin function for small values of $g\mu_B S(-AM+B)/kT$, the magnetization M is approximated as

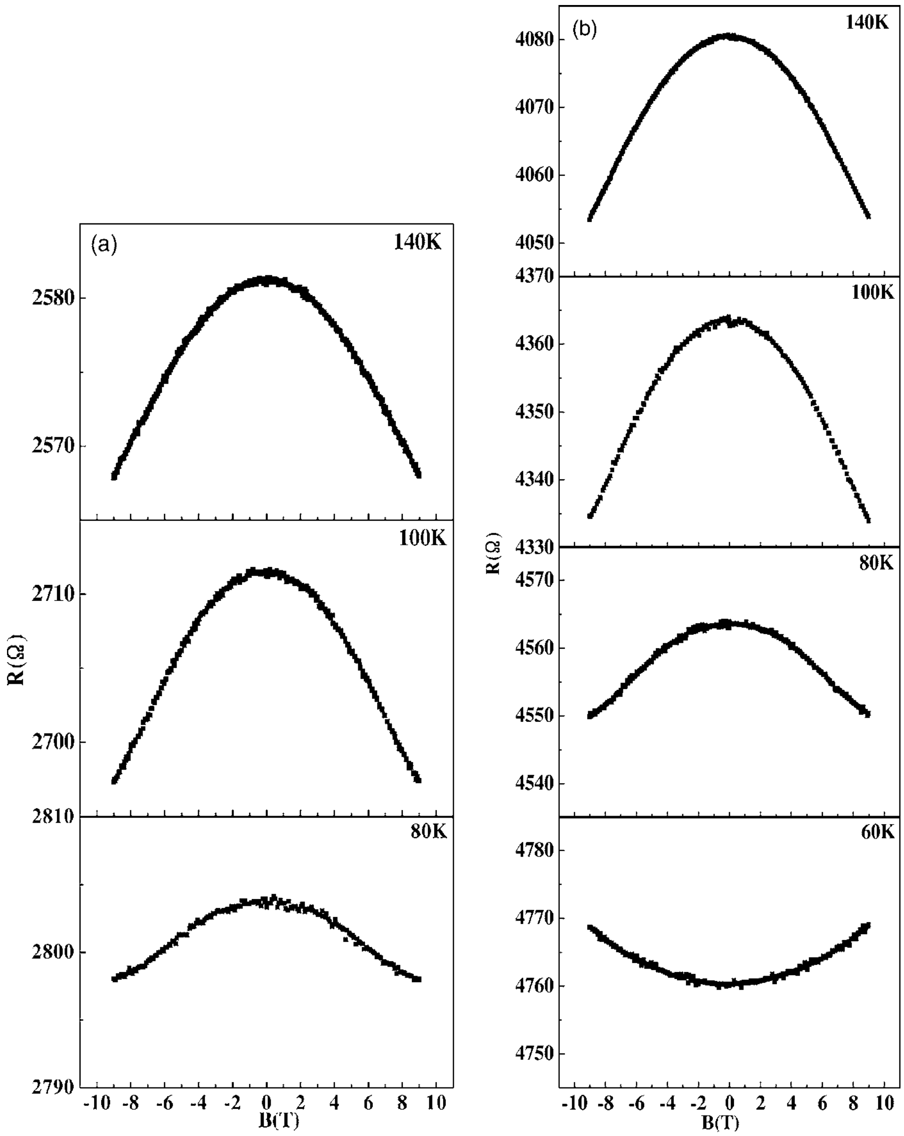

FIG. 5. Resistance of (a) sample 3 and (b) sample 5 as a function of the magnetic field which was applied in the direction parallel to the current and δ-doped layers.

$$M = N(T)g\mu_B S\left\{\frac{S+1}{3S}\left[\frac{Sg\mu_B B}{k(T+T_0)}\right] - \frac{[(S+1)^2+S^2](S+1)}{90S^3}\left[\frac{Sg\mu_B B}{k(T+T_0)}\right]^3\right\}, \quad (6)$$

where

$$T_0 = \frac{2}{3k}S(S+1)\left\langle \sum_m J(R_m)\right\rangle. \quad (7)$$

Here, $\langle \Sigma_m J(R_m)\rangle$ is the average value for the remaining free spins.

On the basis on Eqs. (1) and (6), Hall resistance can be written as a function of the temperature T and magnetic field B in the following form:

$$R_{Hall} = R_H^0 B + N(T)\left[A_1\frac{B}{(T+T_0)} - A_2\frac{B^3}{(T+T_0)^3}\right], \quad (8)$$

where A_1 and A_2 are constants. By using this equation, Hall resistance curves corresponding to those of sample 3 in Fig. 4(a) were calculated. In the calculations, T_0 was assumed to be constant, although it should vary with the temperature according to the above definition. The normal term $R_H^0 B$ of Hall resistance was assumed to be equal to the Hall resistance at 50 K, R_{Hall}(50 K), for all other temperatures. Two parameters, $N(T)A_1$ and $N(T)A_2$, were first determined by fitting a calculated curve to the experimental curve observed at 180 K for a given value of T_0. Next, the value of T_0 was determined by fitting shapes of the curves calculated with the above two parameters to those of experimental curves for 140 and 100 K. Finally, the ratios N(140 K)/N(180 K) and N(100 K)/N(180 K) were determined by fitting magnitudes of the calculated resistance to the experimental resistance for these two temperatures. In Fig. 6, calculated curves obtained in this way are plotted with experimental curves of $R_{Hall}(T) - R_{Hall}$(50 K). The best fit was obtained with T_0=150 K, N(140 K)/N(180 K)=0.88, and N(100 K)/N(180 K)=0.45. In Fig. 6, calculated curves obtained by assuming T_0=0 and constant N, which correspond to a noninteracting spin system, are also plotted. These calculations show that experimental Hall resistance curves can be reproduced fairly well

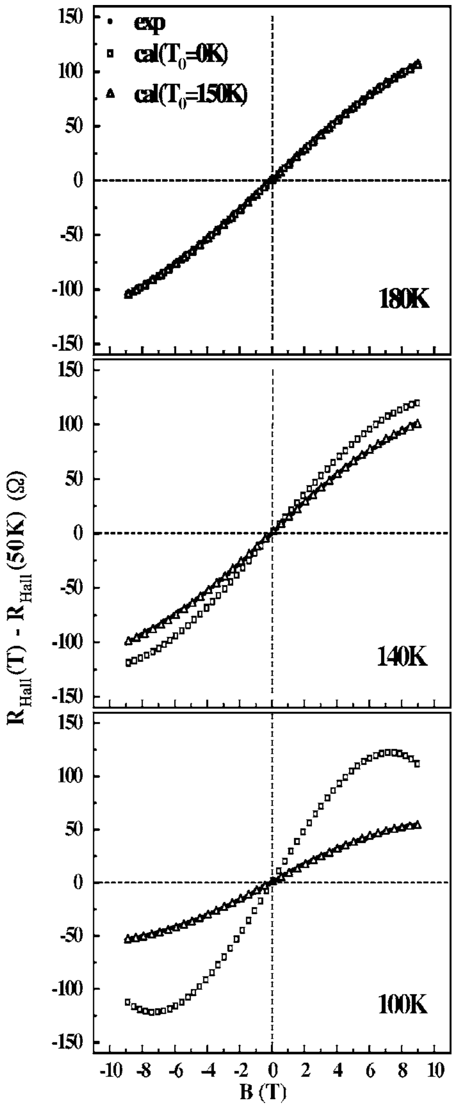

FIG. 6. Experimental and calculated curves of $R_{Hall}(T) - R_{Hall}(50\text{ K})$ for sample 3. Curves denoted by exp are experimental curves. Curves denoted by cal(T_0=0 K) were calculated with $T_0=0$ K and the temperature-independent N. Curves denoted by cal(T_0=150 K) were calculated with $T_0=150$ K and N which decreased with lowering the temperature.

on the basis of an antiferromagnetically interacting spin system.

By using Eqs. (1) and (6), a concentration of localized magnetic moments at 180 K was estimated with parameters determined in the above calculation. From the ratio A_2/A_1, a set of values of S and g was first estimated by using Eq. (6). One example of such sets is $S=3/2$ and $g=40$. With these values of S and g, $N(180\text{ K})$ was calculated by using the values of ρ_{Hall}, R_H^0, and ρ in Figs. 2 and 3(b), which were obtained from a van der Pauw sample. For this calculation, it was assumed that $n=1$ and $c \approx 1.0\text{ T}^{-1}$ in Eq. (1) as in the case of the anomalous Hall effect of $Ga_{1-x}Mn_xAs$.[18] The estimated value of $N(180\text{ K})$ is $8\times10^{19}\text{ cm}^{-3}$, which corresponds to a concentration of localized magnetic moments in the current-flow region at 180 K. If the width of the current-flow region is assumed to be 3 nm, the sheet concentration of localized magnetic moments becomes $2.4\times10^{11}\text{ cm}^{-2}$, which is approximately 1% of the uncompensated Be concentration of this sample. Although we assumed a value of the constant c similar to that of $Ga_{1-x}Mn_xAs$ and used a particular set of values of S and g which allow only a crude estimation, the order of the estimated value suggests that the model considered here reasonably describes the underlying process of the observed results. The g value estimated above is very large in comparison with those of localized spins in impurity-doped semiconductors. If a small value is chosen for g, a large value of S has to be assumed in order to account for the observed Hall resistance curves. In an earlier electron-spin-resonance study on acceptor states in GaAs,[19] large g values were also estimated from the observed results. Along with these earlier results, the present result may suggest a unique property of localized spins associated with acceptor states in GaAs.

In the past, possible freezing of localized magnetic moments and spin-glass formation in impurity-doped semiconductors were investigated by a number of experimental studies.[7,20] These studies showed that freezing of an entire localized magnetic moment system did not occur down to the measurable lowest temperatures. A theoretical study showed that these experimental observations could be explained on the basis of an intrinsic nature of a highly random antiferromagnetic system where the exchange coupling varied in a broad range due to random arrangements of impurity atoms.[21] The explanation of the present results on the basis of the freezing hence appears to contradict with the results of these past studies. There is, however, one significant difference of the pair δ-doped structure from impurity-doped bulk semiconductors with respect to the distribution of impurity atoms. A scanning-tunneling-microscope (STM) study on Be δ-doped layers with high doping concentrations showed that Be atoms in δ-doped layers formed short-range-ordered arrangements, which were ascribed to Coulombic repulsive interactions among them.[22] The Be concentrations in our samples are comparable to those used in the STM study, and hence such short-range-ordered arrangements of Be atoms are expected to have occurred in these samples. Antiferromagnetic ordering of localized magnetic moments may have occurred in the Be δ-doped layers because of nearly equal interatomic spacings of Be atoms, resulting in complete freezing of moments at low temperatures.

On the basis of the above analyses, it is reasonable to consider the existence of localized magnetic moments in the δ-doped structure at high temperatures. It is unlikely that the close correlation among four sets of results shown in Figs. 3(a), 3(b), 4, and 5 is caused by artifacts resulting from measurement processes or occurs accidentally. It is also difficult to find other mechanisms with which one can explain the observed correlation in a consistent manner. The most important result which enables one to elucidate the origin of the correlation among the four sets of results is the nonlinear dependence of the Hall voltage on the magnetic field in Fig. 4. This nonlinear dependence directly corresponds to the Brillouin function which represents the thermal average of localized magnetic moments in a paramagnetic state.

There is one question with respect to the present results which needs to be clarified at the next stage of the study.

There is a large variation of the magnitude of negative magnetoresistance among metallic samples which were grown under similar doping conditions; samples 2 and 4 show small negative magnetoresistance in comparison to samples 3 and 5. As seen in Figs. 3(a) and 3(b), the temperature dependence of R_H/ρ and magnetoresistance of samples 2 and 4 are similar to those of sample 1 which has a single Be δ-doped layer. This suggests that the majority of holes in samples 2 and 4 are in extended states with fewer holes being in localized states like the case of sample 1. In samples 3 and 5, on the other hand, more holes are considered to be localized, as indicated by a larger increase of their sheet resistance at lower temperatures in Fig. 2; at low temperatures, the number of holes excited thermally to extended states is reduced. We found that negative magnetoresistance and R_H/ρ significantly vary among other metallic samples with similar doping conditions, while these values are always close to one another among insulating samples grown under similar doping conditions.[10] A ratio of concentrations of itinerant and localized holes in a metallic sample, hence, seems to change sensitively with slight differences of MBE growth conditions. One such condition may be a ratio of Be and Si doping concentrations, and the other the substrate temperature; the precise control of the substrate temperature among different MBE growth experiments is known to be difficult. It is speculated that slight differences of these conditions result in a change of impurity distributions and hence potential profiles in the pair δ-doped structure, which then affect a ratio of concentrations of itinerant and localized holes.

In summary, metallic samples of the Be and Si pair δ-doped GaAs structure have been studied by combining Hall resistance and magnetoresistance measurements. The anomalous Hall resistance and negative magnetoresistance were observed from these samples at high temperatures. Dependence of anomalous Hall resistance on the temperature and applied magnetic field is closely correlated to that of negative magnetoresistance for each sample. Their dependence is explained on the basis of a paramagnetic state of interacting localized magnetic moments coexisting with itinerant holes in the samples. These results strongly suggest the existence of localized magnetic moments in the pair δ-doped structure at high temperatures.

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