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# Cooperative transition of electronic states of antisite As defects in Be-doped low-temperature-grown GaAs layers

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# Cooperative transition of electronic states of antisite As defects in Be-doped low-temperature-grown GaAs layers

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Magnetic properties resulting from localized spins associated with antisite arsenic ions $As_{Ga}^{+}$ in Be-doped low-temperature-grown GaAs (LT-GaAs) layers were studied by measuring the magnetization of lift-off samples. With fast cooling, the magnetization of samples at 1.8 K becomes significantly lower than that expected from Curie-type paramagnetism in the range of the applied field to 7 T, and a transition from low magnetization to the magnetization of paramagnetism occurs upon the heating of samples to 4.5 K. With slow cooling, on the other hand, samples have a paramagnetic temperature dependence throughout the measurement-temperature range. The magnetization was found to decrease monotonically when a sample was kept at a fixed low temperature. These observations are explained by the cooperative transition of electron states of $As_{Ga}$ defects, which is closely related to the normal-metastable state transition of EL2 defects in semi-insulating GaAs. The results of the magnetization measurements in the present study suggest that $As_{Ga}^{+}$ ions are spontaneously displaced at low temperature without photoexcitation in Be-doped LT-GaAs. The similarity of the transition observed in this system to the normal-metastable state transition of the EL2 defect was also suggested by first-principle calculations of the electron state of an $As_{Ga}$ defect with a doped Be atom. © *2011 American Institute of Physics*. [doi:10.1063/1.3671059]

## I. INTRODUCTION

Point defects that introduce deep electronic states in bandgaps of semiconductor crystals have been extensively studied experimentally and theoretically because of the technological importance of understanding their properties.[1–3] In such studies, novel phenomena, such as the metastability of EL2 defects in GaAs and the negative *U* property of DX centers in AlGaAs have stimulated a great deal of interest, since their occurrence is inherent to the fundamental electronic property of point defects in semiconductor crystals. In these defects, there tend to be large atomic displacements due to a change in the chemical bond scheme, and the displacements are accompanied by a change in the electron configuration, resulting in significant changes to the magnetic and optical properties of defects. Large atomic displacements lead to a strain field in a crystal, through which point defects are expected to interact with each other. Interactions via the strain field act over a long range. Therefore, they may lead to a cooperative change in electron configurations in point defects even at relatively low defect concentrations. The occurrence of such a cooperative phenomenon has not been reported to date. It is, however, important to investigate such a possibility because its realization is expected to present a fundamentally interesting problem in the field of semiconductor physics.

In this paper, we present results of a study on localized spins in GaAs layers grown at low temperatures (LT-GaAs) by molecular-beam epitaxy (MBE), which indicate the cooperative transition of electronic states of antisite As ($As_{Ga}$) defects. A LT-GaAs layer contains a high concentration of $As_{Ga}$ defects up to $1 \times 10^{20}$ $cm^{-3}$.[4] The electronic structure of an $As_{Ga}$ defect has been investigated in many theoretical studies in connection with EL2 defects in semi-insulating (SI) GaAs,[5–12] which are known to undergo a normal-metastable state transition.[13] It has been shown that the $As_{Ga}$ defect induced three antibonding states of $A_1$ symmetry, among which one fell deep in the bandgap and was a twofold occupation in the neutral state.[5,6,9,11] The occupation of the antibonding state by electrons results in large displacements of adjacent As atoms away from the $As_{Ga}$ atom.[9] In undoped LT-GaAs, a few $As_{Ga}$ atoms changed into $As_{Ga}^{+}$ ions, owing to the compensation by Ga vacancies. These $As_{Ga}^{+}$ ions carry localized spins associated with unpaired *sp*-type electrons, which were detected in a number of past studies with electron paramagnetic resonance[14] and the magnetic circular dichroism of absorption.[15] In earlier studies on LT-GaAs, the concentration of $As_{Ga}^{+}$ ions was found to increase when doping a high concentration of Be atoms, which are shallow acceptors and, hence, compensate for $As_{Ga}$ atoms instead of Ga vacancies.[16,17] In recent studies, we grew thick Be-doped LT-GaAs layers with thicknesses up to 20 $\mu$m and measured their magnetic moments with a super-conducting quantum interference device (SQUID).[18,19] From the analysis of the field dependence and temperature dependence of the magnetization of the layers, we found weak antiferromagnetic interactions of localized spins resulting from their direct exchange interaction.

In the present study, we grew Be-doped LT-GaAs layers whose $As_{Ga}^{+}$ concentrations are considerably higher than

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those of samples investigated in the earlier studies[18,19] and measured their magnetization by changing the condition of cooling and field application. In a fast cooling sequence, the magnetization of samples at 1.8 K was found to be significantly lower than that expected from Curie-type paramagnetism, and a transition from low magnetization to that of paramagnetism occurs upon heating a sample to 4.5 K. These observations are explained by the cooperative transition of electronic states of $As_{Ga}$ defects, which is closely related to the normal-metastable state transition of EL2 defects in SI GaAs. The normal-metastable state transition of EL2 defects is now widely considered to result from the displacement of a neutral $As_{Ga}$ atom from the tetrahedral site to an interstitial site via photoexcitation.[13] The magnetization measurements of Be-doped LT-GaAs in the present study suggest that $As_{Ga}^{+}$ ions are spontaneously displaced at low temperature without photoexcitation. First-principle calculations of the electron state of an $As_{Ga}$ defect with a doped Be atom in a GaAs crystal further suggest the possible similarity of the transition observed in this system to the normal-metastable state transition of EL2 defects.

## II. EXPERIMENTAL

Beryllium-doped LT-GaAs layers were grown, employing a conventional MBE system with semi-insulating epiready (100)GaAs substrates that were cut into 15 mm $\times$ 18 mm rectangles. After desorption of an oxide layer of the substrate surface, an undoped 150-nm-thick GaAs buffer layer was grown at 580 °C. For the samples to be used for magnetization measurements, an AlAs layer and a 75-nm-thick GaAs buffer layer were grown at 580 °C; the AlAs layer was made for the lift-off of LT-GaAs layers.[20] The substrate temperature was subsequently lowered for the growth of a Be-doped LT-GaAs layer. Growth procedures were explained in detail in earlier reports.[18,19]

Table I lists substrate temperatures $T_s$, Be concentrations [Be], thicknesses $t$, and spin concentrations $N_s$ of four samples in magnetization measurements. The four samples are divided into two groups: samples A and B have high spin concentrations, while samples C and D have low spin concentrations. The spin concentrations were estimated from magnetization data, as explained later.

Samples A and B were grown at substrate temperatures lower than those for samples C and D. Reflection high-energy electron diffraction patterns of the former two samples were found to change around a layer thickness of 5 $\mu$m, indicating an increase in surface roughness, while such a change was not observed during the growth of samples C and D up to 18 $\mu$m and 15 $\mu$m, respectively. The increase in the surface roughness is known to lead eventually to the formation of extended defects in a LT-GaAs layer.[21,22] For each of sample A and sample B, therefore, two pieces with thicknesses equal to or less than 5 $\mu$m were grown under identical conditions to obtain sufficient magnetic moments for the magnetization measurement. Thicknesses of the samples are listed in Table I. Rocking curves of these samples observed by x ray diffraction are similar to curves presented in an earlier report,[18] indicating high crystalline quality without extended defects.

The magnetization of the four samples was measured using a Magnetic Property Measurement System SQUID system from Quantum Design. For the measurement, each LT-GaAs layer was cut into a $3 \times 3\,\text{mm}^2$ square and lifted from a GaAs substrate by etching the AlAs layer with a solution of HF acid to avoid the GaAs substrate and metal indium contributing to the measured magnetic moments, where the metal indium is attached to the backside of the substrate so that the substrate can be fixed to an MBE sample holder. Lift-off samples were wrapped in thin plastic film and installed in a straw used as a sample holder of the SQUID. To avoid inclusion of a small piece of magnetic material, the lift-off process and installation of a sample in the SQUID sample holder were carefully carried out with plastic tweezers.

TABLE I. List of substrate temperatures $T_s$, Be concentrations [Be], thickness $t$, and spin concentrations $N_s$ of four samples used for magnetization measurements.

| Sample | $T_s$ (°C) | [Be] ($10^{19}$/cm$^3$) | $t$ ($\mu$m) | [$N_s$] ($10^{19}$/cm$^3$) |
|---|---|---|---|---|
| A | 240 | 3.0 | 5 | 2.76 |
| | | | 4 | |
| B | 250 | 3.2 | 5 | 2.62 |
| | | | 5 | |
| C | 260 | 2.4 | 18 | 1.79 |
| D | 280 | 2.7 | 15 | 1.40 |

## III. RESULTS AND DISCUSSION

### A. Magnetization

Magnetization results for sample A are presented first as an example. Figure 1 shows the field dependence of the magnetization (M-H curve) of sample A at selected temperatures. As shown in the earlier report,[18] the magnetic moment of the sample holder with a plastic film is nearly independent of temperature, as expected from their diamagnetism, and its magnitude is comparable to that of the temperature-dependent magnetic moment of the Be-doped LT-GaAs layer at low temperatures. To remove the contribution of the sample holder and the plastic film to measured magnetic moments, Fig. 1 plots the difference in the magnetization at a temperature $T$, $M(T)$ and that at 100 K, $M(100\text{K})$. In Fig. 1(a), the field dependence of the magnetization was measured at selected temperatures in the order of 100, 15, 4.5, and 1.8 K, while it was measured in the order of 100, 20, 15, 10, 4.5, 3.5, 2.5, and 1.8 K in Fig. 1(b). In both measurements, the sample temperature was decreased at a rate of 10 K/min in the temperature range from 100 to 15 K and at a rate of 0.5 K/min from 15 to 1.8 K. Figures 1(a) and 1(b) also show magnetization curves calculated on the basis of Curie paramagnetism using the equation

$$M(T) = N_s S g \mu_B B_s\left(\frac{S g \mu_B H}{k_B T}\right), \quad (1)$$

where $B_s(x)$ is the Brillouin function. In the equation, $N_s$, $S$, $g$, $\mu_B$, and $H$ are the spin concentration, spin, Landé $g$ factor,

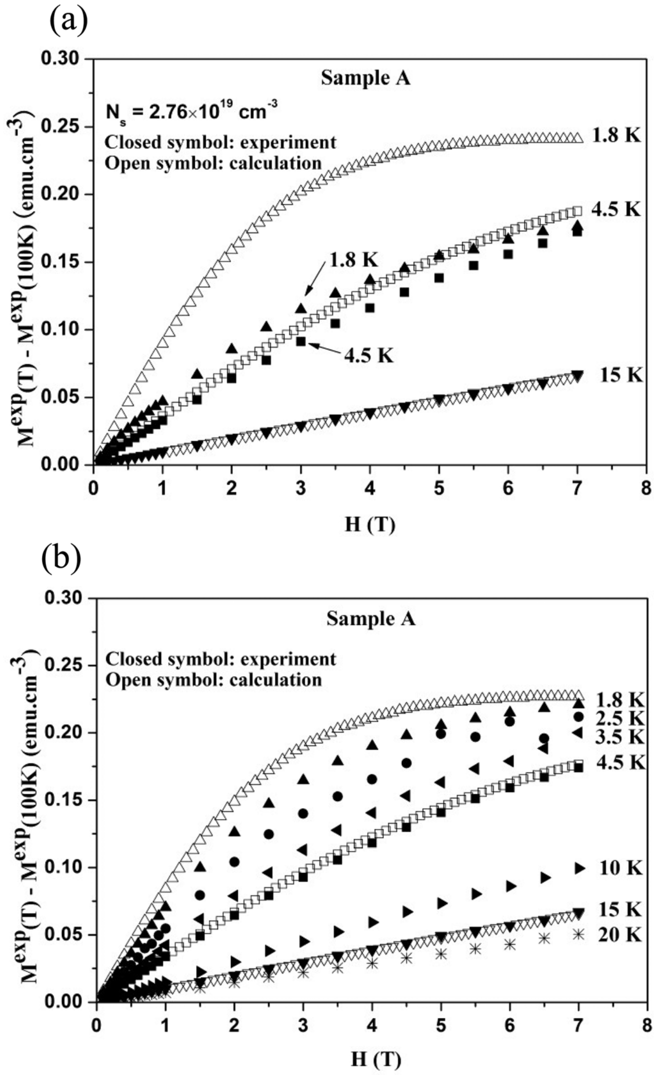


FIG. 1. Field dependence of the magnetization of sample A at selected temperatures for different cooling sequences: (a) 100−15−4.5−1.8 K and (b) 100−20−15−10−4.5−3.5−2.5–1.8 K. Both figures include M-H curves calculated according to Curie paramagnetism and assuming the $g$-factor and spin $S$ are 2.0 and 1/2, respectively.

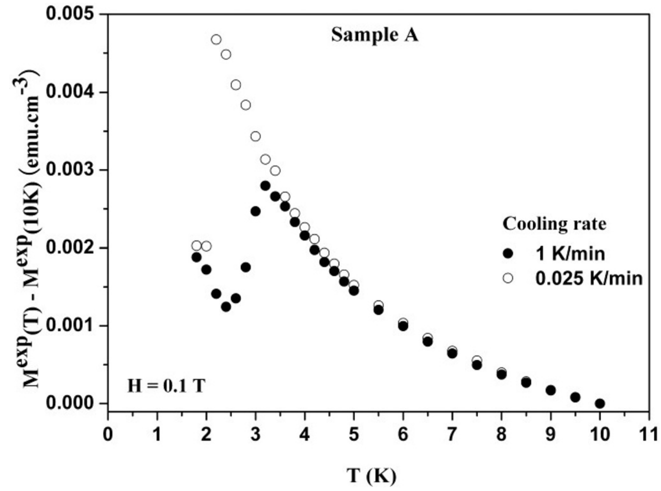


FIG. 2. Temperature dependence of the magnetization of sample A with two different cooling rates.

Bohr magneton, and magnetic field, respectively. Curie paramagnetism implies that each $As^{+}_{Ga}$ ion has a fixed localized spin and the ions do not interact. Our earlier study on the magnetic properties of Be-doped LT-GaAs layers showed that the dependence of the magnetic properties on the applied field and temperature is approximately described by Curie paramagnetism, with a small deviation due to weak antiferromagnetic interactions among localized spins.[19] The calculations were carried out by assuming values of the $g$-factor and the spin $S$ as 2.0 and 1/2, respectively, which were estimated in an earlier electron paramagnetic resonance study on LT-GaAs.[14] The spin concentration $N_s$ was estimated to be $2.76 \times 10^{19}$ cm$^{-3}$ from the measured magnetization at 15 K.

In Fig. 1(a), the measured magnetization at 1.8 K is significantly lower than that expected from Curie paramagnetism. In Fig. 1(b), on the other hand, the measured magnetization at low temperatures, including that at 1.8 K, is close to that expected from Curie paramagnetism. These results indicate that the magnetization at 1.8 K decreases as the sample is cooled, with fewer steps of the M-H curve measurement. For the measurements at 1.8 K, the sample temperature was monitored with a thermometer in the SQUID system and found to be close to 1.8 K in the cases of both Figs. 1(a) and 1(b).

Figure 2 presents the results for measurements 1 and 2 of the temperature dependence of the magnetization of sample A with different cooling rates. For both measurements, the sample was cooled from 300 to 1.8 K without applying a field, and the magnetization was measured upon heating the sample in a magnetic field of 0.1 T. For measurement 1, the cooling rate was set at 10 K/min from 300 to 10 K and at 1 K/min from 10 to 1.8 K, with a 5 min pause at 4.5 K. For measurement 2, the cooling sequence was identical to that of measurement 1, except that the cooling rate was only 0.025 K/min from 10 to 1.8 K. The difference in magnetization at $T$ and 10 K is plotted in the figures. In both measurements, the magnetization at 1.8 K was lower than that expected from Curie paramagnetism, for which magnetic susceptibility changes in proportion to $1/T$. In measurement 1, the magnetization increased in a nearly discontinuous manner at temperatures around 3 K and then monotonically decreased, as in the case of Curie paramagnetism. In measurement 2, on the other hand, there was an abrupt increase in magnetization at 2.2 K, which was considerably lower than the corresponding temperature in measurement 1. These results again show the sensitive dependence of the magnetization of the sample at low temperatures on the sample cooling process.

The results presented in Figs. 1 and 2 imply that the magnetization at 1.8 K was significantly lower than that expected from Curie paramagnetism when the sample was cooled with fewer steps of the M-H curve measurements or without an M-H curve measurement. The measurement of an M-H curve up to 7 T at one temperature takes 120 min. Therefore, it took significantly longer to set a sample at 1.8 K in the measurement of Fig. 1(b) than in the measurements of Figs. 1(a) and 2. Hence, there is the possibility that the low-measured magnetization at 1.8 K in the latter cases may be due to the actual sample temperature differing from

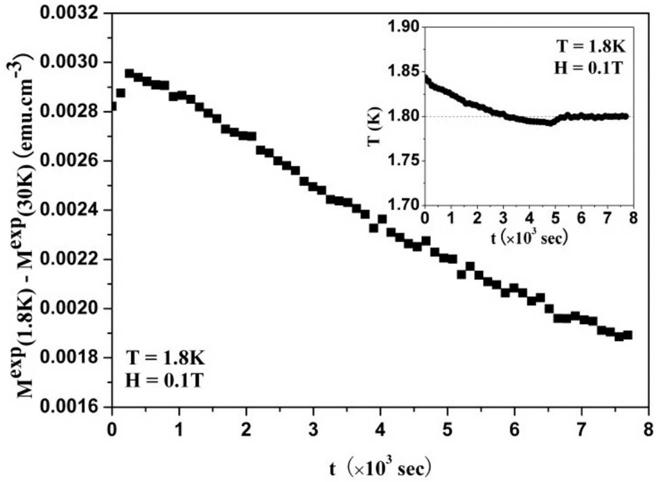


FIG. 3. Magnetization of sample A at 1.8 K in a 0.1-T field as a function of time, where the monitored sample temperature is shown in the inset.

that monitored by the thermometer; the sample temperature may have been higher than the temperature measured by the thermometer, owing to low thermal conduction inside the sample chamber. To examine this possibility, we measured the magnetization of sample A at 1.8 K in a field of 0.1 T as a function of time. The sample was cooled to 1.8 K in the same way as for measurement 1 in Fig. 2. Figure 3 shows the result of the measurement, where the monitored sample temperature is shown in the inset. The magnetization slightly increased, owing to a relatively rapid decrease in the temperature in the initial stage, but decreased monotonically afterward. The continuous decrease in magnetization at a fixed temperature in Fig. 3 implies that the low-measured magnetization at 1.8 K does not result from the sample temperature being higher than the monitored temperature. Instead, the result indicates that the continuous decrease in magnetization with time is due to a gradual change in the sample toward the equilibrium state.

Figures 4(a), 4(b), and 4(c), respectively, show the temperature dependence of the magnetization of samples B, C, and D for two different cooling rates. Spin concentrations $N_s$ of the three samples, which were estimated by comparing the measured M-H curves at 15 K with the calculated ones, are listed in Table I. The measurement processes, including the sample cooling condition, are the same as those for the results shown in Fig. 2. These results for samples B, C, and D were similar to those for sample A, exhibiting different transition behaviors for different cooling rates. There was, however, an important difference in the results between samples with high spin concentrations and those with low spin concentrations. In the case of samples A and B, the magnetization at 1.8 K for a cooling rate of 0.025 K/min was significantly lower than that expected from Curie paramagnetism and comparable to that for a cooling rate of 1 K/min. In the case of samples C and D, on the other hand, the magnetization at 1.8 K for the cooling rate of 0.025 K/min was slightly lower than that expected from Curie paramagnetism. Figures 2 and 4 also show that the temperature of the transition from low magnetization to high magnetization for the cooling rate of 1 K/min was higher for a sample with a higher spin concentration; the temperatures of peak magnetization for samples A, B, C, and D were 3.2, 3.0, 2.8, and 2.4 K, respectively. These results indicate that a sample with higher spin concentration has greater tendency to shift to a low-magnetization state, which in turn is more stable against sample heating.

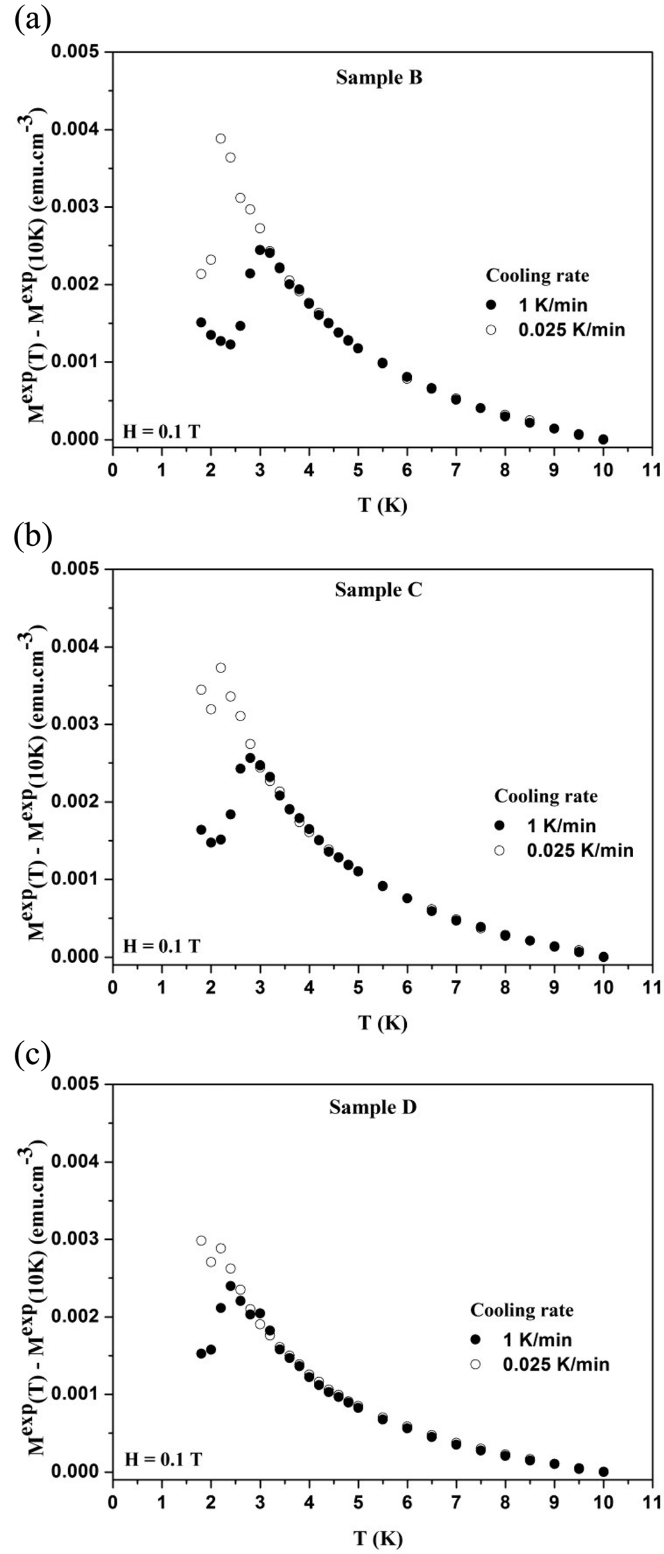


FIG. 4. Temperature dependence of the magnetization for (a) sample B, (b) sample C, and (c) sample D at different cooling rates.

As shown by the results presented above, Be-doped LT-GaAs samples have low magnetization at low temperatures under certain cooling conditions. These observations cannot be attributed to the inclusion of small pieces of other materials, such as ferromagnetic materials and metal indium in the sample holder. In the case of the latter possibility, metal indium becomes superconducting around 3 K in weak magnetic fields of a few hundred Gauss and has a large negative magnetic moment.[23] With the sensitive dependence of the occurrence of low magnetization on the condition of cooling and field application and the persistence of low magnetization in the whole measurement range to 7 T, one can exclude this possibility. As explained earlier, the low measured magnetization at 1.8 K cannot be attributed to a difference in the actual sample temperature from the monitored temperature.

In our earlier study, we showed that localized spins associated with $As_{Ga}^{+}$ ions in Be-doped LT-GaAs antiferromagnetically interact with each other via direct exchange.[19] One may attribute the low magnetization at low temperature to the freezing of these localized spins resulting from the antiferromagnetic interactions. The sensitive dependence of the occurrence of low magnetization on the sample cooling condition and field application may be considered to result from the frustration of interaction due to disordered arrangements of localized spins, similar to the case for spin glasses.[24] The persistence of the low magnetization up to 7 T and its occurrence in samples with low spin concentrations at temperatures around 3 K, however, make such a possibility highly unlikely.

With the elimination of the above-mentioned possibilities, we consider the most likely origin of the transition observed in the present study to be a change in the electronic state of $As_{Ga}^{+}$ ions, which is closely related to the normal-metastable state transition of EL2 defects in SI-GaAs. EL2 defects in SI-GaAs transform to the metastable state at low temperature under the irradiation of photons with energy ranging from 1.0 to 1.3 eV, and the recovery to the normal state occurs upon heating the crystal at about 120 K.[13] The EL2 defect is a deep donor in its normal state, while it becomes electrically and optically inactive in the metastable state. It is now well established that the EL2 defect is an isolated $As_{Ga}$ atom and the normal-metastable state transition occurs via the displacement of a neutral $As_{Ga}$ atom between the tetrahedral site and an interstitial site.[9,13] Additionally, earlier studies showed that $As_{Ga}$ atoms in LT-GaAs undergo normal-metastable state transition.[25–28] With a transition to the metastable state, $As_{Ga}$ is no longer a deep donor and is thus expected to become a neutral defect without localized spin. The transition can occur even in a sample with a low concentration of $As_{Ga}$, as in the case of the EL2 defect in SI-GaAs.

### B. First-principle calculation

In Sec. III A, the displacement of an $As_{Ga}$ defect to an interstitial site was considered the most likely origin of the observed transition from high magnetization to low magnetization at low temperature. Although a number of first-principle calculations have been carried out for the transition of a neutral $As_{Ga}$ atom from the normal state to the metastable state,[5–12] no calculation has been reported for the transition of an $As_{Ga}$ atom in the presence of a shallow acceptor. Therefore, we carried out first-principle calculations for the transition of an $As_{Ga}$ atom with a Be atom in a GaAs crystal to investigate the change in the spin distribution.

A GaAs $(2 \times 2 \times 2)$ supercell with size of 11.306 Å was used for the calculations. One $As_{Ga}$ atom was placed at the center of the supercell, while one Be atom occupied the second-nearest-neighbor cation site of the $As_{Ga}$ atom. For the metastable state, the $As_{Ga}$ atom was initially displaced in the [111] direction by 1.2 Å, which was obtained in an earlier theoretical study on a neutral $As_{Ga}$ atom in the metastable state.[9] All configurations were then fully relaxed to reach the minimum-energy structures. Density functional theory was used for the calculations.[29,30] The generalized gradient approximation functional in the Perdew-Burke-Ernzerhof formula developed by Perdew, Burke, and Ernzerhof for the exchange correlation and double numerical plus polarization basis set were used.[31] A $4 \times 4 \times 4$ Monkhorst-Pack grid of k-points was employed for the Brillouin-zone integrations. All calculations were performed using Dmol$^3$ code.[32]

Figure 5 presents the spatial spin density distribution, density of states (DOS), and spin DOS of two models: (a) a Be atom and an $As_{Ga}$ atom at substitutional sites in a GaAs crystal and (b) a Be atom at a substitutional site and an $As_{Ga}$ atom at an interstitial site in a GaAs crystal. For each DOS and spin DOS, the energy $E = 0$ corresponds to the Fermi energy. To observe the spin density distribution in each model, calculations were made by imposing the condition that the total spin is conserved in each supercell; antiferromagnetic coupling between spins in adjacent supercells was not allowed.

In Fig. 5(a), there is a highly localized spin distribution around the $As_{Ga}$ site, and its state is located in the middle of the bandgap, as seen in the DOS and spin DOS. In Fig. 5(b), on the other hand, spins are delocalized over all As sites, but there is a tendency for localization around the displaced $As_{Ga}$ atom to a certain degree. As an important difference from Fig. 5(a), the spin state is completely mixed with the valence band in the DOS and spin DOS of model (b). This mixing implies the coalescence of the impurity band made of the states of Be and displaced $As_{Ga}$ atoms with the valence band due to their high concentrations resulting from the small size of the supercell. If a large supercell is used, these states are expected to be located at levels close to the valence band. The large delocalization of spins in Fig. 5(b) also suggests a strong tendency of antiferromagnetic coupling of spins in a real crystal, even at low spin concentrations. Therefore, the results presented in Fig. 5 indicate that localized spins are associated with $As_{Ga}$ ions at substitutional sites, but disappear with the displacement of $As_{Ga}$ ions to interstitial sites at a finite temperature. These results strongly support our explanation of the observed transition in Be-doped LT-GaAs as being similar to the normal-metastable state transition of EL2 defects.

Unlike the transition of EL2 defects in SI-GaAs, the observed transition from high magnetization to low magnetization occurs without light illumination. This implies that the $As_{Ga}$ atom at an interstitial site has lower energy than

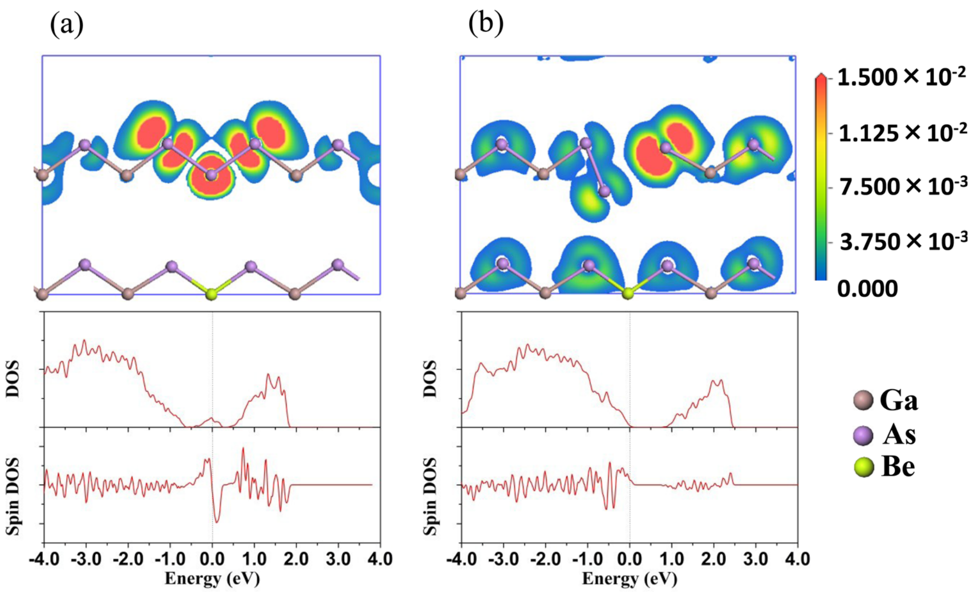


FIG. 5. (Color online) Spatial spin density distribution, density of states (DOS), and spin DOS of two models: (a) a Be atom and an $As_{Ga}$ atom at substitutional sites in a GaAs crystal and (b) a Be atom at a substitutional site and an $As_{Ga}$ atom at an interstitial site in a GaAs crystal. For each DOS and spin DOS, the energy $E = 0$ corresponds to the Fermi energy.

that at a substitutional site at low temperature, resulting in a spontaneous transition. The former state is hence called the displaced state instead of the metastable state in the present paper. As indicated by the results in Figs. 2 and 4, samples with higher spin concentrations and, hence, higher $As_{Ga}^{+}$ concentrations have an increasing tendency to shift to the low-magnetization state at low temperature. This suggests that the energy of the displaced state is lower than that of the normal state because of the mutual interaction of $As_{Ga}$ defects.

As stated in Sec. I, the most interesting possibility suggested by the present results is a cooperative nature of the transition of $As_{Ga}$ defects, which would result from interactions among the defects. The two following results suggest the cooperative nature of the transition. The first result is the nearly abrupt transition from the low magnetization state to the high magnetization state in Figs. 2 and 4. If cooperative interactions do not exist, the transition is expected to occur gradually with a change in the temperature because of a gradual change in the Boltzmann distribution of the two states of individual defects.[33] The second result is the sensitive dependence of the transition on the process of cooling a sample, which is seen in Figs. 1, 2, and 4. It is difficult to ascribe this phenomenon to behaviors of individual $As_{Ga}$ defects.

There are peculiar phenomena related to the observed transition. As shown in Figs. 2 and 4, the transition from high magnetization to low magnetization occurs at low temperature, even with the slow cooling of a sample, but the temperature of the reverse transition from low magnetization to high magnetization upon heating depends on the rate of the earlier cooling of the sample. Another peculiar phenomenon is the extremely slow decrease in the magnetization with time at 1.8 K, seen in Fig. 3. The decrease continued over 2 h without giving any indication of slowing. To clarify the underlying microscopic processes of these phenomena, we need to carry out further experimental investigations of microscopic processes of the transition, in particular, the nature of interactions of $As_{Ga}$ defects, employing other methods, such as x ray diffraction analyses, in addition to magnetization measurements.

## IV. CONCLUSION

Our observations indicate a transition of the magnetic state in Be-doped LT-GaAs layers. The observations are explained by the cooperative transition of electron states of $As_{Ga}$ defects, which is closely related to the normal-metastable state transition of EL2 defects in SI-GaAs. The results suggest that $As_{Ga}^{+}$ ions spontaneously transform into the displaced state at low temperature and return to the normal state upon heating. The above explanation of the observed transition is supported by first-principle calculations of electron states of an $As_{Ga}$ defect with a doped Be atom. The occurrence of the transition sensitively depends on the sample cooling condition. It was also found that the low magnetization state continuously developed at a fixed low temperature. These results suggest novel aspects of the transition kinetics resulting from mutual interactions of point defects in a semiconductor crystal.

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