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Description	

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Non-equilibrium critical point in Be-doped low-temperature-grown GaAs

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We studied the transition process of antisite arsenic defects in Be-doped low-temperature-grown GaAs layers by measuring the magnetization. This material exhibits bistability at non-equilibrium; at a fixed temperature in a fixed magnetic field a sample relaxes towards two different states, depending on the preceding cooling process. We observed anomalously large magnetization fluctuations in macroscopic samples during the transition from bistability to monostability with gradual change of the temperature. Slowing down of the relaxation of the magnetization is observed as a sample approaches the transition into monostability. Large fluctuations observed from a two-piece sample exhibit intermittent bursts by high-pass filtering and follow a generalized Gumbel probability density distribution. These observations suggest a possibility of the occurrence of a non-equilibrium critical point in this material. Microscopic processes underlying the observed phenomena are discussed with results of first-principles calculations of strain fields. © 2013 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4790313>]

I. INTRODUCTION

Critical phenomena in non-equilibrium systems are currently a fundamental research subject for broad fields of physics.^{1–3} In contrast to critical phenomena in equilibrium systems, the understanding of the underlying mechanism of non-equilibrium critical phenomena is still limited. This situation is attributed to the lack of a thermodynamic free energy framework and their occurrence in a large variety of physical, chemical, biological, and social systems. There is another significant reason; it is the difficulty in finding experimental systems with which non-equilibrium critical phenomena can be studied under highly controlled conditions. For example, critical exponents of non-equilibrium phase transitions belonging to the directed percolation class have been investigated by many theoretical and simulation studies in past decades,² but the critical behavior of directed percolation was confirmed experimentally for the first time only a few years ago.⁴ In view of this situation, it is important to search for experimental systems that exhibit non-equilibrium critical phenomena and enable us to investigate them under highly controlled conditions. Studies on such systems will provide us with an opportunity of gaining new insights into non-equilibrium critical phenomena.

In a recent study, we found a cooperative transition of electronic states of antisite As(As_{Ga}) defects in Be-doped low-temperature-grown GaAs (LT-GaAs).⁵ This transition results from displacements of As_{Ga}⁺ ions between substitutional sites and interstitial sites. An As_{Ga} defect, known as the EL2 defect, acts as a deep double-donor at a substitutional site, while at an interstitial site it becomes electronically inactive.^{6,7} With the presence of a shallow acceptor Be atom, therefore, the former state has a localized spin by

giving one electron to the Be atom, while the latter does not.⁵ Thus, one can directly monitor the change of As_{Ga} defects in a sample by measuring its magnetization. The unique aspect of this transition is that it occurs through cooperative interactions of point defects in a crystal as indicated by the direct dependence of the transition temperature on the concentration of defects.⁵ Molecular-beam epitaxial (MBE) growth at a low temperature leads to a high concentration of As_{Ga} defects in a GaAs layer which is far greater than the solubility limit of excess As at equilibrium.⁸ Because of the cooperative change of point defects introduced excessively into a crystal, this transition is essentially a non-equilibrium cooperative phenomenon in a condensed matter system.

We have investigated the possibility of critical phenomena in the transition of As_{Ga} defects in Be-doped LT-GaAs layers. This material exhibits bistability in the magnetization at non-equilibrium; at a fixed temperature in a fixed magnetic field, a sample relaxes towards two different states, depending on the prior cooling process. We observed anomalously large fluctuations in the magnetization during the transition from bistability to monostability with gradual change of temperature. Large fluctuations observed from a two-piece sample exhibit intermittent bursts by high-pass filtering and follow an asymmetric non-Gaussian distribution. Slowing down of the relaxation of the magnetization is observed as a sample approaches the transition into monostability. These observations suggest a possibility of the occurrence of a non-equilibrium critical point in the transition. An important implication of these results is that one may be able to study non-equilibrium critical phenomena by direct observation of critical fluctuations in macroscopic samples. In this paper, we present results of the magnetization measurements of Be-doped LT-GaAs layers with the main focus on novel aspects of large fluctuations, and discuss the microscopic processes underlying the observed phenomena with results of first-principles calculations of strain fields.

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II. EXPERIMENTAL

Beryllium-doped LT-GaAs layers were grown with a conventional MBE system with semi-insulating epitaxially (100)GaAs substrates that were cut into a size of 15 mm $\times$ 18 mm. After desorption of an oxide layer of the substrate surface, an undoped 150-nm-thick GaAs buffer layer was grown at 580 °C. To use the samples for magnetization measurements, we grew an AlAs layer and a 75 nm-thick GaAs buffer layer at 580 °C; the AlAs layer was made for the lift-off of LT-GaAs layers.⁹ The substrate temperature was subsequently lowered for the growth of a Be-doped LT-GaAs layer. The growth procedures and sample preparation method for the magnetization measurement are explained in detail in earlier reports.^{5,11,12}

Table I lists substrate temperatures T_s , Be concentrations [Be], thicknesses t , and spin concentrations N_s of two samples used in the present magnetization measurements. We estimated the spin concentrations from magnetization data, as explained later. Sample A was grown at a substrate temperature lower than that of sample B. We found a change in the reflection high energy electron diffraction pattern of the former sample around a layer thickness of 5 μ m, indicating an increase in the surface roughness, but did not observe such a change during the growth of sample B up to 18 μ m. The increase in the surface roughness is known to lead to the formation of extended defects in a LT-GaAs layer.¹² Therefore, for sample A, two pieces with thicknesses equal to or less than 5 μ m were grown under identical conditions to obtain sufficient magnetic moments for the magnetization measurement. Sample thicknesses are listed in Table I.

We measured the magnetization of two samples with a MPMS SQUID system from Quantum Design. For the measurement, each LT-GaAs layer was cut into a size of 3 $\times$ 3 mm² and lifted off a GaAs substrate by etching the AlAs layer with a solution of HF acid to avoid the GaAs substrate and metal indium contributing to measured magnetic moments. The metal indium was attached to the backside of the substrate so that the substrate could be fixed to a MBE sample holder. Lift-off samples were wrapped in a thin plastic film and installed in a straw used as a SQUID sample holder. We put sixty 3 $\times$ 3 mm² pieces and two 3 $\times$ 3 mm² piece in the sample holder for sample A and sample B, respectively. For each measured value of the magnetic moment, the scan of a sample through the detection coil was repeated for ten times, and the signals were averaged to improve signal-to-noise ratio.

TABLE I. List of substrate temperature T_s , Be concentration [Be], thickness t , spin concentration N_s , and quantity of lift-off pieces used for magnetization measurements.

Sample	T_s (°C)	[Be] (10 ¹⁹ /cm ³)	t (μ m)	N_s (10 ¹⁹ /cm ³)	Quantity (piece)
A	240	3.0	5	2.76	60
B	260	2.5	18	1.95	2

III. RESULTS

A. Bistability

Fig. 1(a) shows two sets of the temperature-dependence of the magnetization of sample A. One set is denoted as a fast cooling sequence and the other a slow cooling sequence. We obtained the results of the fast cooling sequence upon heating in a magnetic field of 100 mT after cooling the sample from 300 to 10 K at a rate of 10 K/min and from 10 to 1.8 K at 1 K/min without applying a field. We obtained the results of the slow cooling sequence after keeping the sample at 10 K for 2 h and cooled to 1.8 K at a rate of 0.025 K/min. The measurement was made upon heating at 100 mT, similarly to the former case. The difference in magnetization at a temperature T and 10 K is plotted for each set. Subtracting $M^{\text{exp}}(10\text{ K})$ removes the temperature-independent contribution of the plastic film and sample holder to the measured magnetic moment.¹⁰

The temperature-dependence of the magnetization for the slow cooling sequence is similar to that of Curie-type paramagnetism in the whole measurement temperature range. For the fast cooling sequence, on the other hand, the magnetization changes abruptly from low to high around 2.6 K. This change is ascribed to cooperative displacements of As_{Ga}⁺ ions

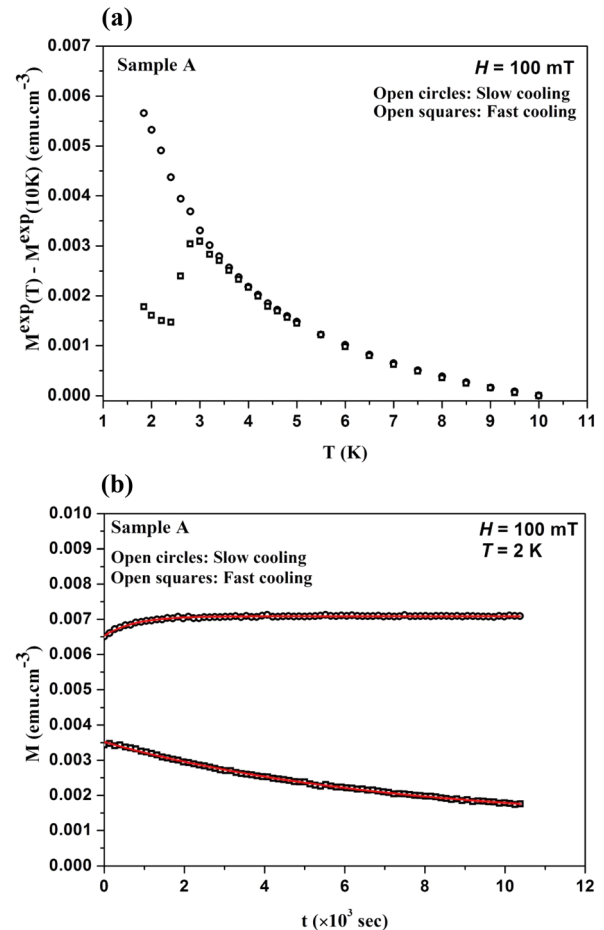


FIG. 1. (a) Temperature dependence of magnetization of sample A for slow cooling and fast cooling. (b) Magnetization of sample A at 2.0 K in a magnetic field of 100 mT as a function of time. Solid lines were derived by the least-square fit of Eq. (2).

from interstitial sites to substitutional sites.⁵ In this paper, we name the former and latter states the displaced and normal states, respectively. These results, hence, indicate that nearly all regions of the sample remain in the normal state in the slow cooling sequence, while a substantial part of the sample changes into the displaced state in the fast cooling sequence. From the temperature dependence of the magnetization in the high temperature range from 4 to 10 K, we estimated the spin concentration N_s listed in Table I using the formula of Curie-type paramagnetism

$$M(T) = \frac{N_s s(s+1)g^2 \mu_B^2 B}{3k_B T}, \quad (1)$$

where $s = 1/2$ and $g = 2.0$.¹³

Fig. 1(b) shows the magnetization of sample A at a constant temperature of 2.0 K in a magnetic field of 100 mT as a function of time. The results shown by open circles were obtained after the sample was cooled to 2.0 K via the slow cooling sequence used for the measurement in Fig. 1(a). Those shown by open squares, on the other hand, were obtained after the sample was cooled to 2.0 K via the fast cooling sequence. The zero-value of the magnetization in the figure is extrapolated using Eq. (1) for infinitely high temperature. The magnetization M , hence, corresponds to the magnetic moment of localized spins in a unit volume of a LT-GaAs sample and is plotted this way in the rest of our figures.

The results in Fig. 1(b) show that the sample approaches different states for both cases under the fixed temperature and applied field. We compare changes in the magnetization for both cases with the relaxation formula

$$M(t) = M(\infty) + \Delta M \exp(-t/\tau), \quad (2)$$

where ΔM is a positive and negative constant for fast and slow cooling, respectively, and τ is the relaxation time. The solid lines in the figure were obtained with Eq. (2). These lines represent both changes fairly well except for the early part of the fast cooling where the sample temperature was not fully stabilized because of the fast cooling from high temperatures. The value of τ is 742 ± 23 s for slow cooling and 7150 ± 180 s for fast cooling. The results in Fig. 1(b) indicate bistability at non-equilibrium. Depending on a sample cooling condition from high temperature, the system falls into the basin of one of two stable states, namely, the normal state and displaced state, and then relaxes to the bottom of the basin.

B. Transition from bistability to monostability

In order to investigate the transition process from bistability to monostability, we measured the magnetization of a sample by gradually changing the temperature after fast cooling. Fig. 2(a) shows results of the measurement of sample A when the temperature was changed stepwise from 2.0 to 3.2 K as shown in Fig. 2(b). The sample was kept at each temperature for 4 h in an applied field of 100 mT. The figure shows that the magnetization continuously decreased at 2.0, 2.4, and 2.8 K and suddenly increased during the change from 2.8 to

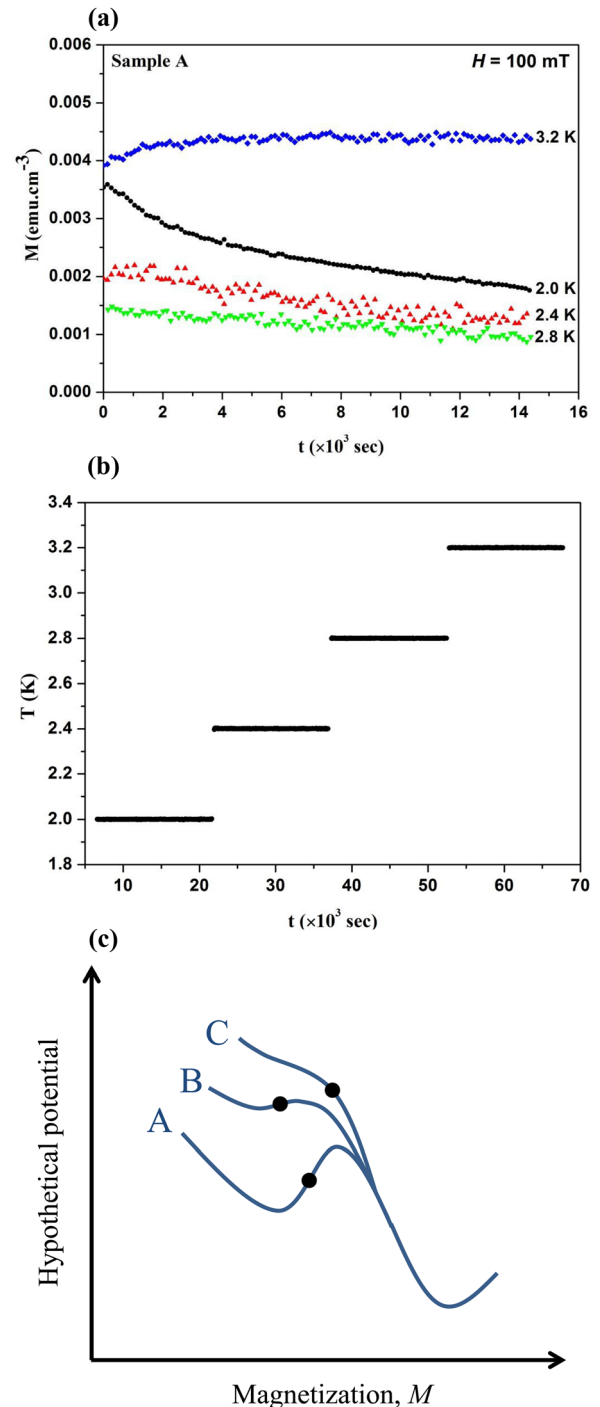


FIG. 2. (a) Change in magnetization of sample A after fast cooling in which the temperature was changed stepwise from 2.0 to 3.2 K. (b) Change of measured sample temperature. (c) Hypothetical potential distributions illustrating the transition from bistability to monostability.

3.2 K. The magnetization exhibits anomalously large fluctuations at 2.4 K and 2.8 K, while it smoothly changes at 2.0 K and has relatively small fluctuations at 3.2 K. The measured sample temperature was stable at each temperature as seen in Fig. 2(b), implying that large fluctuations spontaneously occur in the sample. Fluctuations at 2.4 and 2.8 K are on the order of $1 \times 10^{-4} \text{ emu} \cdot \text{cm}^{-3}$, which corresponds to several percentages of the magnetization at these temperatures.

Fig. 2(c) is hypothetical potential distributions that illustrate the situations in Fig. 2(a). Potential distributions

A, B, and C correspond to the situations at 2.0 K, 2.4 K (2.8 K), and 3.2 K, respectively, where a dot indicates the location of the system. As shown in this figure, potential distribution B is close to a situation corresponding to the transition point from bistability to monostability. We repeated this measurement for six times for sample A and obtained similar results.

Spontaneous large fluctuations suggest that the sample may approach a critical state where the spatial correlation diverges. To investigate this possibility, we analyzed a change of the relaxation rate by least-squares fit of Eq. (2) to the data of the above-mentioned measurements. Fig. 3(a) is the fit to the data of the measurement at 2.4 and 2.8 K in Fig. 2(a). For 2.4 K, a broken line was obtained by the fit of Eq. (2) with a relaxation time of $11\,980 \pm 2960$ s. For 2.8 K, however, the least-squares fit of Eq. (2) could not be made due to extremely large standard errors of estimated parameters. Solid lines in the figure are explained in Sec. IV.

In Fig. 3(b), the average value of relaxation times estimated by the fit of Eq. (2) to data of six measurements is plotted as a function of the temperature. For each of 2.4 and 2.8 K, data of four measurements were included because of the inability of the least-squares fit to data of the other two measurements. Although measurements were made only for three temperatures, the figure clearly indicates an increase of

the relaxation time as the sample approaches the transition into monostability, suggesting the critical slowing down of the relaxation. The relaxation times derived from the data in Fig. 1(b) for fast cooling are significantly longer than that for slow cooling. This long relaxation time in the former case suggests that the slowing down has already started at 2.0 K.

Fluctuations at 2.4 and 2.8 K in Figs. 2(a) and 3(a) occur at fast rates, while the relaxation is very slow at these temperatures. This difference in the time scale is explained as follows. Sample A consists of 60 pieces, and each piece is divided into a number of statistically independent domains. In individual domains, As_{Ga} defects collectively change between the normal and displaced states at fast rates. On the other hand, the slow magnetization relaxation described by Eq. (2) emerges as a collective property of temporal variations of the number of As_{Ga} defects in the normal state in all individual domains of 60 pieces, which are considered as mutually independent stochastic processes.

In Figs. 2(a) and 3(a), amplitudes of fluctuations in the magnetization at 2.4 K are greater than those at 2.8 K. This difference is attributed to the Curie-type paramagnetism. An As_{Ga} defect in the normal state possesses a localized spin, while that in the displaced state does not. Magnetic interactions between localized spins in Be-doped LT-GaAs layers are very weak as shown by an earlier study.¹¹ The magnetic moment of a sample hence is inversely proportional to the temperature for a given number of As_{Ga} defects in the normal state, resulting in larger fluctuations in the magnetization at a lower temperature for a given size of fluctuations in the number of As_{Ga} defects in the normal state.

C. Non-Gaussian fluctuations

The sample holder contained 60 pieces of lift-off sample A for measuring the magnetization. These 60 pieces contribute independently to the observed magnetic moments. In order to investigate intrinsic properties of the large fluctuations, we measured the magnetization of sample B that contains only two lift-off pieces. Fig. 4(a) is the temporal dependence of magnetization fluctuations of sample B at 2.0, 2.4, and 2.8 K which was measured under the same condition with that of Fig. 2(a). As expected, the fluctuations in the magnetization of sample B are several times larger than those of sample A. Because only two lift-off pieces were included in the sample holder, measured magnetic moments of sample B are significantly small. The figure shows an error bar corresponding to the standard deviation due to noises for this measurement condition which was estimated by measuring a reference sample. As seen in the figure, fluctuations due to measurement noises are not significant in comparison to spontaneous fluctuations of the sample.

Figs. 4(b) and 4(c) show the same signals as in Fig. 4(a) at 2.8 K and those subjected to high-pass filtering with the filtering frequency 2.5×10^{-3} Hz, respectively. High-pass filtered signals show intermittent bursts similar to those observed in velocity fluctuations of turbulent fluids with high-pass filtering.¹⁴ As the filtering frequency is increased, signals become increasingly intermittent. Such a tendency suggests that observed fluctuations are not Gaussian.¹⁴

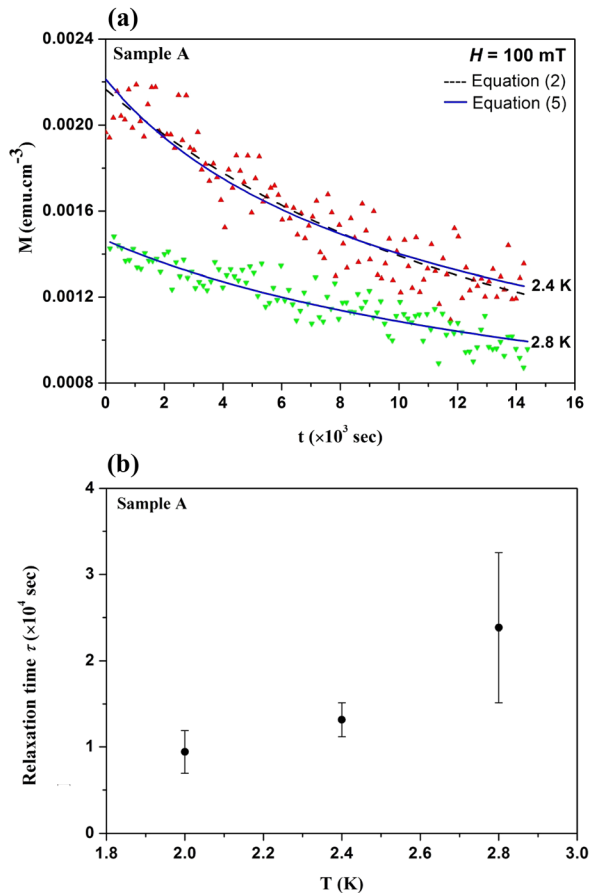


FIG. 3. (a) Magnetization fluctuation of sample A at 2.4 and 2.8 K with a broken line and solid lines obtained by the least-squares fit of Eqs. (2) and (5), respectively. (b) Relaxation time τ of sample A derived by the least-squares fit of Eq. (2) as a function of temperature.

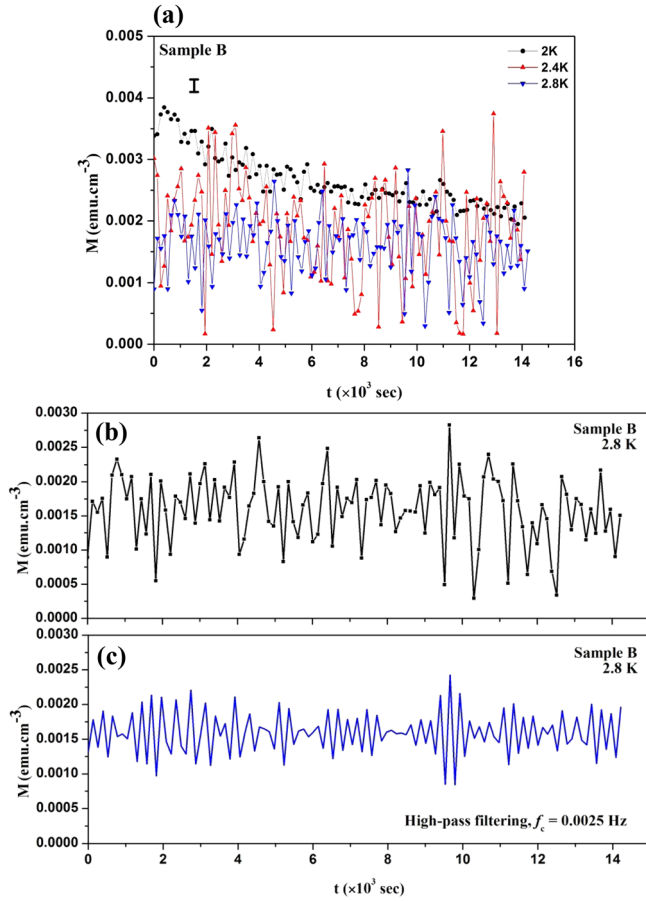


FIG. 4. (a) Temporal dependence of magnetization fluctuations of sample B at 2.0, 2.4, and 2.8 K which was measured under the same condition with that of Fig. 2(a). (b) The same signals as in Fig. 4(a) at 2.8 K and (c) those subjected to high-pass filtering with the filtering frequency 2.5×10^{-3} Hz.

To obtain the distribution of fluctuations, a curve of the form of Eq. (2) was fitted to the data of each measurement at one temperature. Numbers N of the deviation ΔM from this curve are plotted in histograms for 2.8 K in Fig. 5(a). We include the data of three measurement sequences. The distributions shown in Fig. 5(a) are skewed towards the negative side and have large deviations on both positive and negative sides. Such skewness and large deviations are also seen directly in fluctuations shown in Fig. 4(b). A similar distribution is observed in fluctuations at 2.4 K.

Fig. 5(b) plots normalized probabilities $\sigma P(\Delta M/\sigma)$ made from histograms for 2.8 K, where σ is the standard deviation. In the figure, the Gaussian probability density function (PDF) is shown by a blue line. Normalized probability densities deviate systematically from the Gaussian PDF. Bramwell *et al.* showed that PDFs of the global quantities take similar non-Gaussian forms in a number of highly correlated systems.¹⁵ The non-Gaussian form is represented by the generalized Gumbel (GG) PDF

$$\sigma P(x) = w \exp[ab(x-s) - ae^{b(x-s)}], \quad (3)$$

where parameters w , b , and s depend on the parameter a . In Fig. 5(b), a GG PDF is shown by a red line with the parameter a of 5.1. The experimental data are close to the GG PDF except for a large deviation on the positive side. According

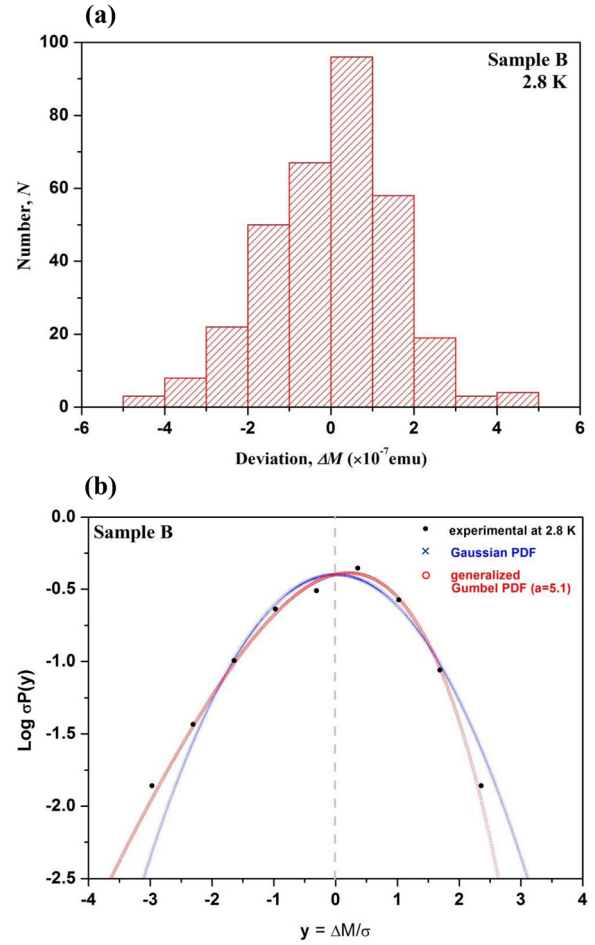


FIG. 5. (a) Histogram of numbers N of the deviation ΔM for 2.8 K. (b) Normalized probabilities $\sigma P(\Delta M/\sigma)$ made from histograms for 2.8 K with the Gaussian PDF and GG PDF.

to the analysis of the low-temperature phase of the two-dimensional XY model,¹⁶ the parameter a is related to a number of statistically independent domains N_{eff} by

$$a \sim \frac{\pi}{2} \left[1 + \frac{N_{\text{eff}}}{4\pi^2} \right]. \quad (4)$$

With Eq. (4), the value $a = 5.1$ implies that sample B is divided into roughly 140 statistically independent domains.

IV. DISCUSSION

Theoretical studies on non-equilibrium states of a macroscopic system^{17,18} have suggested that there is a critical point in the transition from bistability to monostability at non-equilibrium. In these studies, the time evolution of a macro-variable is assumed to follow stochastic master equations with a WKB-like approximation¹⁷ or with a power-series expansion.¹⁸ The first and second derivatives of the first moment of the transition probability vanish at a steady state corresponding to a critical point. These studies suggest that the enhancement of fluctuations, slowing down of the relaxation, and non-Gaussian distributions of fluctuations occur as a system approaches the critical point. The observation of anomalously large fluctuations of macroscopic

samples, slowing down of the relaxation, and non-Gaussian properties of fluctuations, therefore, suggests that there is a non-equilibrium critical point in Be-doped LT-GaAs. An important implication of the finding in the present study is that one may be able to study non-equilibrium critical phenomena by direct observation of critical fluctuations in macroscopic samples. In recent years, GG-like non-Gaussian distributions have attracted a great deal of interest as they occur in a variety of highly correlated systems.^{15,19–23} Hence, the study on this material will provide us with an opportunity of experimentally investigating this important subject.

In the theoretical study referred above,¹⁷ Kubo *et al.* derived an equation representing the slowing down of the relaxation in the neighborhood of the critical point

$$M(t) = \frac{\Delta M}{(1 + 2|\gamma_3|\Delta M^2 t)^{1/2}}, \quad (5)$$

where γ_3 is the third derivative of the first moment of the transition probability. A similar equation was derived by van Kampen.¹⁸ Both equations lead to an asymptotic relaxation as $t^{-1/2}$. The least-squares fit of Eq. (5) to the data of 2.4 and 2.8 K leads to solid lines in Fig. 3(a) with estimated parameters $2|\gamma_3|\Delta M^2 = (1.49 \pm 0.10) \times 10^{-4} \text{s}^{-1}$ and $2|\gamma_3|\Delta M^2 = (8.16 \pm 0.53) \times 10^{-5} \text{s}^{-1}$ for 2.4 and 2.8 K, respectively. As seen in the figure, both lines derived by the fit of Eqs. (2) and (5) are close to each other for 2.4 K, but the standard error of parameters determined by the least-squares fit of Eq. (5) is smaller than that of Eq. (2), which further suggests the possibility of critical slowing down.

In order to obtain more convincing evidence for the occurrence of the criticality in this system, however, we need to observe an unambiguous power-law relation in this system. For such an investigation, we need to identify a control parameter with which a critical point is quantitatively defined. Temperature appears to be such a parameter in the measurement presented in Sec. III B, but the critical point is not determined solely by temperature in the present case as

explained below. It is necessary to fully clarify microscopic processes of the transition in order to identify a control parameter and derive power-law relations.

With the experimental and theoretical results which have been obtained up to the present for this material, we consider microscopic processes underlying the bistability and large fluctuations as follows. According to the first-principles calculation,⁷ the total energy of an As_{Ga} defect in the displaced state is higher than that in the normal state by 0.24 eV. Therefore, a sample with a very low concentration of As_{Ga} defects is expected to relax monotonically towards the normal state at a fixed temperature. Interactions among As_{Ga} defects, however, are expected to modify the relaxation process significantly in a sample with a relatively high concentration of defects. A change of the structure of an As_{Ga} defect between the normal and displaced state is expected to result in a large strain in the surrounding lattice. We made first-principles calculations of lattice structures for the normal and displaced states. The calculation method is described in our earlier paper,⁵ but we used a larger supercell that contains 27 GaAs cells at this time. With one As_{Ga} and Be atom in a supercell, their concentrations are comparable to those of experimental samples. Figs. 6(a) and 6(b) show (110) planes of the supercell for the normal and displaced states, respectively. Sizes of the deviation of the interatomic spacing $\Delta d/d$ from that in the perfect GaAs crystal, 0.2448 nm, are shown by different colors in the figures. As seen in these figures, a change in $\Delta d/d$ on the order from 10^{-2} to 10^{-3} occurs over the nearly whole supercell when the As_{Ga} atom transforms from the normal state to the displaced state. Many As_{Ga} defects, therefore, collectively contribute to the strain field and lead to long range elastic interactions among them. Another important structural aspect of As_{Ga} defects is their random distribution in a crystal. A high-energy and complex strain field, therefore, is expected to form if both normal and displaced states coexist in a sample. Such a strain field suppresses transitions of As_{Ga} defects from the displaced state to the normal state if the latter is the minority state in a sample; a number of As_{Ga}

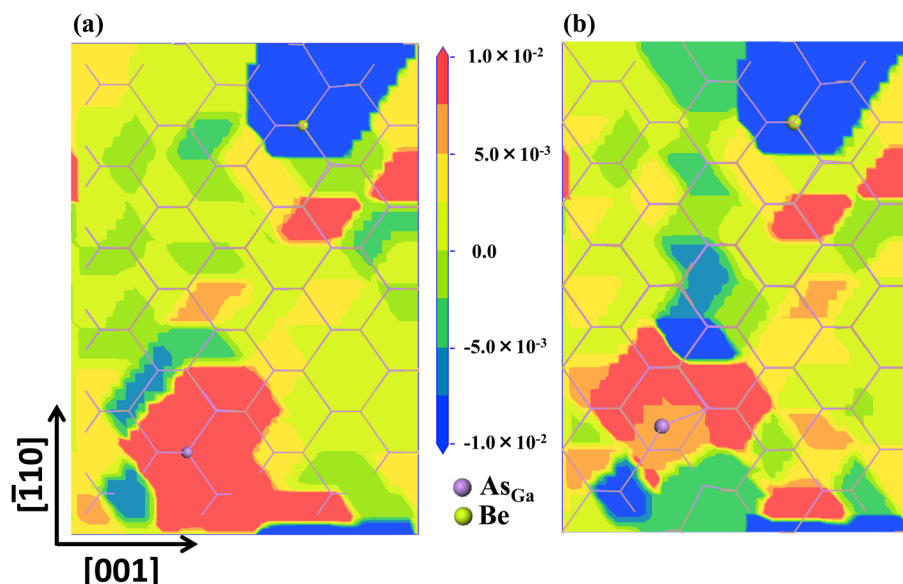


FIG. 6. (110) planes of the supercell containing an As_{Ga} and Be atom for (a) the normal state and (b) displaced state, showing sizes of the deviation of the interatomic spacing $\Delta d/d$ from that in the perfect GaAs crystal.

defects in the normal state decreases so as to remove a complex strain field, resulting in bistability at non-equilibrium.

With a slow increase in the temperature of a sample in which both normal and displaced states coexist, the average phonon energy becomes barely comparable to the elastic energy due to the coexistence of two states at a certain temperature and then induces chain reactions of As_{Ga} defects over the whole sample with successive changes of the complex strain field. If the temperature is not sufficiently high as in the case of 2.4 and 2.8 K, chain reactions of As_{Ga} defects continue to occur towards either normal or displaced state, resulting in sustained large fluctuations. The occurrence of the critical state, therefore, depends on the temperature, but it also directly depends on the strain field caused by the coexistence of two states which is affected by the preceding cooling process of a sample.

In summary, we measured magnetization to study the transition of As_{Ga} defects in Be-doped LT-GaAs layers. This material exhibits bistability at non-equilibrium; at a fixed temperature in a fixed magnetic field, a sample relaxes towards two different states, depending on the prior cooling process. We observed anomalously large fluctuations in the magnetization in macroscopic samples in the transition from bistability to monostability with gradual change of temperature. Slowing down of the relaxation of the magnetization is observed as a sample approaches the transition into monostability. Large fluctuations observed from a two-piece sample exhibit intermittent bursts by high-pass filtering and follow a GG-like non-Gaussian distribution. These observations suggest a possibility of a critical point in this material at non-equilibrium.

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