Magnetic phase diagram of \( \text{Sr}_{2-x}\text{La}_x\text{IrO}_4 \) synthesized by mechanical alloying

K. Horigane,1 M. Fujii,2 H. Okabe,3 K. Kobayashi,1,2 R. Horie,1 H. Ishii,3 Y. F. Liao,4 Y. Kubozono,1 A. Koda,3 R. Kadono,3 and J. Akimitsu1
1Research Institute for Interdisciplinary Science, Okayama University, Okayama, Japan
2Graduate School of Natural Science and Technology, Okayama University, Okayama, Japan
3Institute of Materials Structure Science/J-PARC Center, KEK, Ibaraki, Japan
4National Synchrotron Radiation Research Center, Hsinchu 30076, Taiwan

(Received 10 October 2017; revised manuscript received 23 January 2018; published 28 February 2018)

We report the crystal structure and physical properties of \( \text{Sr}_{2-x}\text{La}_x\text{IrO}_4 \) synthesized by mechanical alloying. The magnetic transition temperature \( T_N \) and electrical resistivity decreased with increasing La doping, consistent with previous studies involving single-crystalline samples. We also identified the relationship between \( T_N \) and tetragonal distortion \((c/a)\) in this system. This result suggests that the magnetism of the \( \text{Sr}214 \) system is strongly correlated with its crystal structure. Zero-field muon spin rotation/relaxation studies revealed that short-range antiferromagnetic ordering is realized in \( \text{Sr}_{2}\text{La}_0\text{IrO}_4 \); also, the spin-glass state is stabilized in the low-temperature region. The Ir moment estimated from the longitudinal field \( \mu\text{SR} \) results is \( 0.045 \mu_B \), indicating that electrons are introduced into the Ir atoms.

DOI: 10.1103/PhysRevB.97.064425

I. INTRODUCTION

Due to their wide variety of physical properties, such as charge and spin ordering, colossal magnetoresistance, and high-\( T_c \) superconductivity, \( 3d \) transition-metal oxides (TMOs) have attracted much attention. These physical properties emerge from the interplay between the spin, charge, and orbital degrees of freedom. In contrast, 5d TMOs should exhibit exotic phenomena because their spin-orbit coupling (SOC) is one order of magnitude larger than that of 3d TMOs. Recently, Kim et al. proposed a spin-orbit Mott state with a local \( J_{\text{eff}} \) \( = 1/2 \) wave function produced by the interplay between SOC and Coulomb \( U \) in \( \text{Sr}_2\text{IrO}_4 \) [1]. In this insulator, which effectively contains one electron per Ir ion, the pseudospin remains a good quantum number and orders antiferromagnetically. Resonant inelastic x-ray scattering (RIXS) revealed that the magnon dispersion in \( \text{Sr}_2\text{IrO}_4 \) is well described by the antiferromagnetic (AF) Heisenberg model with a superexchange interaction of \( J_{\text{eff}} \) \( = 1/2 \) on a square lattice [2]. More recently, our group performed angle-resolved photoelectron spectroscopy (ARPES) measurements of electron-doped \( \text{Sr}_{2-x}\text{La}_x\text{IrO}_4 \) \((x = 0, 0.04, 0.08)\), and observed a dispersive in-gap state that evolves with carrier doping [3]. This in-gap state was also observed in hole-doped \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) [4] and electron-doped \( \text{Nd}_{2-x}\text{Ce}_x\text{CuO}_4 \) [5]. Due to the similar Mott physics between cuprates and iridates, \( \text{Sr}_2\text{IrO}_4 \) is a good candidate for exploring unconventional high-\( T_c \) superconductivity by carrier doping.

Watanabe et al. theoretically predicted that electron-doped \( \text{Sr}_2\text{IrO}_4 \) exhibits \( d \)-wave superconductivity formed by the \( J_{\text{eff}} = 1/2 \) Kramers doublet [6]. Instead, a triplet \( p \)-wave superconducting state was found on the hole-doping side [7]. The physical properties of electron-doped \( \text{Sr}_2\text{IrO}_4 \) systems have been extensively studied in experiments by several groups. Castaneda et al. reported the structure and magnetic properties of polycrystalline samples of \( \text{Sr}_{2-x}\text{La}_x\text{IrO}_4 \). They found that the electrical resistivity increases with increasing La concentration, and their magnetic susceptibility results revealed canted antiferromagnetism below 240 K in both \( \text{Sr}_2\text{IrO}_4 \) and electron-doped \( \text{Sr}_{1.85}\text{La}_{0.15}\text{IrO}_4 \) [8]. However, in neutron scattering and magnetic susceptibility measurements using single-crystalline samples of \( \text{Sr}_{2-x}\text{La}_x\text{IrO}_4 \), the long-range AF order was suppressed up to \( x = 0.08 \) and the short-range AF order persisted up to \( x = 0.12 \) with La doping [9]. Qi et al. reported that introducing oxygen vacancies into single-crystal \( \text{Sr}_2\text{IrO}_4 \) \((0 < \delta < 0.04)\) significantly decreased the resistivity; also, \( \text{Sr}_2\text{IrO}_4 \) \((\delta = 0.04)\) underwent a metal-insulator transition at 105 K [10]. Thus the differences in the physical properties of polycrystal and single-crystal \( \text{Sr}_{2-x}\text{La}_x\text{IrO}_4 \) remain an unresolved issue.

In previous studies of polycrystalline \( \text{Sr}_{2-x}\text{La}_x\text{IrO}_4 \) samples, the powder samples were synthesized by a short heating and quenching technique. Therefore, in this study, we synthesized our samples using mechanical alloying (MA), which is well known to improve chemical reactions. We report the crystal structure and physical properties of \( \text{Sr}_{2-x}\text{La}_x\text{IrO}_4 \) derived from synchrotron powder x-ray diffraction, magnetic susceptibility, electrical resistivity, and muon-spin relaxation (\( \mu\text{SR} \)) measurements.

II. EXPERIMENTAL PROCEDURE

Polycrystalline samples of \( \text{Sr}_{2-x}\text{La}_x\text{IrO}_4 \) were prepared by conventional solid-state reactions. A mixture of \( \text{SrCO}_3 \) \((0 < x < 0.1)\), \( \text{SrO} \) \((0.11 < x < 0.2)\), \( \text{La}_2\text{O}_3 \), and \( \text{IrO}_2 \) was ground and further mixed by planetary ball milling (Fritsch, P-7) at a rotation rate of 400 rpm for 3 h with 15 (5 mm-diameter) and 10 (10 mm-diameter) \( \text{ZrO}_2 \) balls. The resulting powders were calcined in air at 1150°C for 15 min.

Magnetic susceptibility measurements were performed using a superconducting quantum interference device magnetometer (Quantum Design MPMS-R2). The electrical resistivity was measured by a conventional dc four-probe method.
with a measuring current $I$ of 1 mA in the temperature range 2–300 K. Scanning electron microscope (SEM) images were recorded using a field emission SEM (FE-SEM) ULTRA55 scanning electron microscope at 10 kV.

X-ray diffraction studies were performed using a conventional x-ray spectrometer with a graphite monochromator (RINT-1100, Rigaku). Synchrotron powder XRD experiments were conducted using the BL12B2 beamline at SPring-8. The incident beam (wavelength $\lambda = 0.6887$ Å) was focused to $250 \mu m^2$ by a toroidal mirror. The samples were sealed in capillaries (diameter 0.5 mm) under an Ar atmosphere. The diffraction data were analyzed using the Rietveld analysis program RIETAN2000 [11].

Zero-field (ZF)- and longitudinal field (LF)-$\mu$SR experiments were performed using the Advanced Research Targeted Experimental Muon Instrument at the S line (ARTEMIS) spectrometer [12] with a fly-past chamber at the Material and Life Science Experimental Facility (MLF), J-PARC, Tokai, Japan.

III. RESULTS AND DISCUSSION

A. Crystal structure

Figure 1 shows the synchrotron x-ray diffraction patterns of $x = 0$ and 0.1. The diffraction patterns of the La-doped samples indicate a tetragonal structure in the $I4_1/acd$ space group. The final reliable factors ranged from 2.04% to 3.01%. In the $x = 0.2$ sample, a small amount of impurity phase (La$_2$O$_3$) was observed. The lattice parameters and the Ir-O-Ir bond angles of ball-milled $\text{Sr}_2\text{IrO}_4$ are summarized in Fig. 2. The $a$ axis expands with $x$, while the $c$ axis shrinks. The contracted $c$ axis can be explained by the much smaller ionic radius of La$^{3+}$ (1.216 Å) compared to that of Sr$^{2+}$ (1.31 Å). However, this idea is inconsistent with the expanded $a$ axis. As the $a$ axis reflects the Ir-O bond length in the IrO$_2$ plane, its enhancement might be due to electron doping in the Ir-O antibonding orbitals. Indeed, the Ir-O-Ir bond angle increases monotonically with La substitution. To clarify the structural differences between ours and previous results, we studied the results of two earlier reports [8,9]. In a polycrystalline sample [8], the doping dependence of the lattice parameters was similar to our observations, but the quantitative values largely differed from our data. On the other hand, the structural parameters of single-crystalline samples [9] explained both the trend and values of the doping dependence. Thus the structural parameters of the MA samples are quantitatively consistent with the single-crystal sample.

From spatially resolved optical second- and third-harmonic generation rotational anisotropy measurements, Torchinsky et al. recently reported a decrease in symmetry from $I4_1/acd$ to $I4_1/a$ in $\text{Sr}_2\text{IrO}_4$ below $T_N$ [13]. To explore the structural changes below and above $T_N$ (~240 K), we performed synchrotron x-ray measurements from 300 to 100 K. Although we detected no structural changes below $T_N$, we observed negative thermal expansions [Fig. 2(d)]. According to Ranjbar et al. [14], negative thermal expansion of the $c$ axis reflects a structural phase transition from $I4_1/acd$ to $I4_1/mmm$ near 1123 K. Thus this unusual behavior probably originates from the structural phase transition.

B. Resistivity

Figure 3(a) shows the electrical resistivity $\rho$ of $\text{Sr}_2-x\text{La}_x\text{IrO}_4$. All samples ($x = 0, 0.1$, and 0.2) exhibit semiconducting behavior, and $\rho$ at 300 K decreases by two orders of magnitude in the La-doped samples. This behavior agrees with previous reports, including measurements of single crystals [9]. However, the observed results contrast with a previous report discussing the introduction of impurity centers by La doping [8], in which the resistivity increased at 300 K. The decreased resistivity observed here strongly supports that La was successfully doped into $\text{Sr}_2-x\text{La}_x\text{IrO}_4$ in the milling technique.

In a previous report of single-crystal $\text{Sr}_2\text{IrO}_4$, the resistivity did not display simple Arrhenius-type behavior $\rho(T) = \rho_0 \exp(\Delta/k_B T)$ [15], but was well fitted by the three-dimensional variable range hopping (VRH) model $\rho(T) = \rho_0 \exp(\Delta/k_B T)^{1/\nu}$ with $\nu = 1/4$ over a certain temperature range. Similar behavior is depicted in Fig. 3(b). The curve of $x = 0$ fits the aforementioned VRH model below 60 K, while that of $x = 0.1$ shows similar behavior over a wider range (below 100 K). The good fits to the VRH model indicate that the behavior of the carriers in these systems is not simply thermally activated. In contrast, the resistivity at $x = 0.2$ displays Arrhenius-type behavior over the entire temperature range. The calculated gap $\Delta$ changed to 11.5 meV ($x = 0$), 12.0 meV ($x = 0.1$), and 2.7 meV ($x = 0.2$). The decrease in the energy gap suggests that electron carriers are introduced into the IrO$_2$ plane via La substitution. This behavior agrees well with the evolved in-gap state induced by La doping, as observed in ARCES measurements of single-crystalline samples [3], as well as the previous resistivity measurement results using single-crystalline samples [9]. The behavior contradicts the previous report on polycrystalline samples, in which the energy gap $\Delta$ slightly increased from 47 meV ($x = 0$) to 55 meV ($x = 0.2$) after La doping [8].
the result indicates here that the mechanical milling technique enables us to synthesize the crystals of which the electrical transport is not bound by grain boundaries which have been limiting the studies of the materials in polycrystalline samples. We also note that the La doping was applied over a wider range in our experiment than in single-crystal experiments, and was similar to the doping range of polycrystalline samples.

To examine the effects of mechanical milling on the morphology of polycrystalline Sr$_{2-x}$La$_x$IrO$_4$ samples, we characterized these samples by SEM. Figures 4(a) and 4(b) show...
the surface morphologies of the MA-prepared Sr_{1.9}La_{0.1}IrO_{4} and the nonalloyed samples, respectively. Comparing the two samples, there are fewer grains in the MA samples at the same scale length, indicating that a post-MA reaction increased the size of the crystal grains and reduced the number of grain boundaries. Therefore, the resistivity difference between our results and those of previous powder studies might originate from grain size and boundary effects.

C. Magnetic susceptibility

Figures 5(a) and 5(b) shows the magnetic susceptibility of Sr_{2-x}La_{x}IrO_{4}. Note that at low temperature, the magnitude of the saturated magnetic susceptibility decreases with increasing La concentration. La doping has been reported to strongly modulate the Ir-O-Ir bond angles [8,9]; thus the present decrease might originate not only from the carrier doping, but also from the approach of the Ir-O-Ir bond angles toward 180°. Both effects would clearly decrease the $T_N$. In the present study, $T_N$ monotonically decreased from 240 K ($x = 0$) to 114 K ($x = 0.13$).

The above results imply a strong relationship between the crystal structure and its magnetism. Recently, Jackeli and Khalilulin [16] and Liu et al. [17] reported the influences of structural perturbations on spin ordering, in particular on the spin canting angle. They constructed a detailed magnetic phase diagram showing the transition from the canted ground state to different types of collinear magnetic states as a function of the octahedral tetragonal distortion ($c/a$). These theoretical studies strongly suggest that $c/a$ is coupled to the magnetism of the Sr214 system. To experimentally clarify the relationship between the crystal structure and its magnetism, we plotted $T_N$ against $c/a$ in Sr_{2-x}La_{x}IrO_{4}. As shown in Fig. 6, $T_N$ decreased with decreasing $c/a$ in this compound. Perkins et al. calculated significant changes in the isotropic exchange coupling $J$ due to the tetragonal distortion [18]. They found that increasing the tetragonal distortion increases the exchange coupling $J$. In this theoretical calculation, the suppression of $T_N$ by $c/a$ can be explained by the decreased exchange coupling of $J$. Similar $T_N$ vs $c/a$ trends have been observed experimentally in Ca doping [19] and Tb doping [20]. These results suggest that the magnetism of a Sr214 system is strongly correlated with the parameters of its crystal structure, such as $c/a$.

Figure 7 shows the magnetic phase diagram of Sr_{2-x}La_{x}IrO_{4} synthesized by MA and single-crystalline samples [9]. The La dependence of $T_N$ in our results favorably

![FIG. 4. SEM images of Sr_{1.9}La_{0.1}IrO_{4}: (a) with ball milling, (b) without ball milling.](image)
agrees with the results of single-crystalline samples. Previous studies reported a short-range AF order above $x = 0.08$ because they found no magnetic Bragg peak measured by neutron scattering. However, it was mentioned that the detection limit of magnetic scattering is $0.06 \mu_B$ in the Sr214 system. Thus it remains unclear whether a short-range AF above $x = 0.08$ can be observed.

**D. $\mu$SR measurement**

The above uncertainty was clarified in $\mu$SR experiments. $\mu$SR is one of the most effective techniques for detecting short-range magnetic orders and small magnetic moments with extremely high sensitivity ($10^{-2}$–$10^{-3} \mu_B$) [21]. Figure 8(a) shows selected ZF-$\mu$SR time spectra of Sr$_{1-x}$La$_x$IrO$_4$ in the temperature range 3.4–300 K. Nonoscillatory signals are present, indicating the absence of a long-range-magnetic order even below $T_N$ ($\sim 180$ K), as also determined by magnetic susceptibility measurements [see Fig. 5(b)]. The oscillations are not completely absent, but the spectral damping becomes faster with decreasing temperature and the spectrum at 3.4 K shows a shallow “dip” that signals freezing of fluctuating random local fields, where the line shape is close to that of a quasistatic spin-glass state [22].

Recently, Chen et al. reported a low-temperature cusp in zero-field cooling (ZFC) data and a slight decrease in the transition temperature, $T_F$, as the La concentration increased. Specifically, $T_F$ reduced from 13 K for $x = 0.02$ to 9 K for $x = 0.12$ [9]. To further probe the transition at $T_F$, they performed low-temperature ac susceptibility measurements on the $x = 0.02$ sample. They found that the peak of the ZFC shifts to higher temperatures with increasing probe frequency. These results support the existence of a spin-glass state at low temperature in doped Sr$_{2-x}$La$_x$IrO$_4$. The time evolution of the spectra above 14 K can be fitted by the following function:

$$A_Z(t) = A_0 \exp(-\lambda t)^\beta + A_{bg}. \quad (1)$$

Here, $A_0$ and $A_{bg}$ are the positron decay asymmetries (amplitudes) of the sample ($A_0 = 0.2048$) and background ($A_{bg} = 0.0293$, mainly from a silver plate) components, respectively; $\lambda$ is the relaxation rate; and $\beta$ is the stretching exponent. Note that the spectra below 300 K are not easily fitted by a simple exponential function ($\beta = 1$). In $\mu$SR analyses, the stretched exponential function is often used to phenomenologically describe relaxation in disordered systems in which the distribution of the relaxation time broadens. The fitting may deteriorate below 14 K due to mixing of the low-temperature phase, as described later.

Figure 8(b) shows the temperature dependences of $\lambda$ and $\beta$. A three-step decrease appears in $\beta$ as the temperature decreases. As evidenced in the lower panel of Fig. 8(b), $\beta$ begins to decrease from approximately 300 K, suggesting the development of short-range spin correlations between the intralayer Ir ions. Subsequently, a gradual decrease of $\beta$ from 0.8 to 0.6 appears around 180 K, corresponding to $T_N$. Because this behavior is often observed in frustrated antiferromagnets, the magnetic state below $T_N$ was determined as a short-range AF order coexisting with spin fluctuation that is detectable within the muon time scale. This situation can also be explained as a Griffiths phase, in which short-range AF clusters occur and grow upon cooling in the paramagnetic phase. Spin autocorrelation functions in the Griffiths phase can be described by a stretched exponential of the form $\exp(-\lambda t)^\beta$ with $\beta = 0.5$ [23], which is also consistent with our fitting results. In fact, an electronic inhomogeneity has been observed in scanning tunneling microscopy (STM) measurements [9], which is attributed to inhomogeneous carrier distribution. Thus these inhomogeneous spin dynamics may be naturally associated with the intermixing of different magnetic phases such as the Griffiths phase.

As the temperature decreases further (to below 50 K), $\beta$ reaches around 1/3, a typical value in dilute spin-glass systems [24,25]. The relaxation rate $\lambda$ also increases rapidly in this
FIG. 8. (a) Temperature dependence of ZF-μSR time spectra for Sr$_{1.9}$La$_{0.1}$IrO$_4$. Solid lines represent the fitting results using Eq. (1). (b) Temperature dependence of the relaxation rate $\lambda$ and the stretching exponent $\beta$. (c) LF-μSR time spectra at 3.4 K. Solid lines represent the best fit results using Eq. (4).

temperature region [see upper panel of Fig. 8(b)]. This implies a critical slowdown of the spin fluctuation toward a spin-glass transition. Indeed, our magnetic susceptibility data ($\chi = 0.1$) under the ZFC process show a kink at 15 K. As similar behaviors have been reported in previous studies [9], we conclude that Sr$_{1.9}$La$_{0.1}$IrO$_4$ has a magnetic phase transition between the short-range order and the spin-glass state at around 14 K.

To more clearly distinguish the low-temperature spin-glass state, we performed LF-μSR measurements on Sr$_{1.9}$La$_{0.1}$IrO$_4$. By applying an external magnetic field parallel to the initial direction of muon-spin polarization, one can evaluate the size of the static-random field at the muon stopping site in a material. Figure 8(c) shows the LF-μSR time spectra of Sr$_{1.9}$La$_{0.1}$IrO$_4$ at 3.4 K under several LFs (50, 100, 150, 200, and 300 G). As the LF increases, the persisting asymmetry is enhanced under restoration of the initial muon polarization; this is usually called the decoupling effect. The decoupling behavior in Fig. 8(c) clearly indicates that most of the Ir electronic spins are static in this state. When the electronic spins are static, dense, and randomly oriented, the LF muon polarization function for a Gaussian local field becomes

$$G^K(t, H_L) = \frac{2\Delta^2}{\omega_L^2} \left[ 1 - \exp \left( -\frac{\Delta^2 t^2}{2} \right) \cos (\omega_LT) \right] + \frac{2\Delta^2}{\omega_L^2} \int_0^t \exp \left( -\frac{\Delta^2 t^2}{2} \right) \sin (\omega_LT) d\tau, \quad (2)$$

where $\Delta$ is proportional to the root-mean square of the local field distribution (Gaussian linewidth), $H_L$ is the longitudinal field, and $\omega_L = \gamma_p H_L$ ($\gamma_p$: muon gyromagnetic ratio = 2$\pi \times 135.54$ MHz/T). Here, we further investigate the contribution of the spin fluctuation $\nu$ as the influence from the neighboring short-range ordered phase. Based on the strong collision model [26,27], the muon-spin polarization function is given as

$$G^K(t, H_L, \nu) = G^K(t, H_L)\exp(-\nu t) + \nu \int_0^t \exp(-\nu \tau) G^K(t-\tau, H_L, \nu) d\tau. \quad (3)$$

Replacing the first term on the right side of Eq. (1) with this form, we obtain

$$A_{LF}(t, H_L) = A_s G^K(t, H_L, \nu) + A_{bg}. \quad (4)$$

The curves are well fitted by the above function, as shown in Fig. 8(c), by sharing the field-independent parameters, $A_s$, $A_{bg}$, $\Delta$, and $\nu$, the so-called global fit. The parameters were extracted as $\Delta = 4.47(3) \mu$s$^{-1}$ and $\nu = 0.111(3)$ MHz; thus the characteristic field at a muon site was determined as $\Delta/\gamma_p = 52.5(3)$ G, approximately two orders of magnitude larger than that of the nuclear magnetic moment ($\sim 0.5$ G) [28].

Before discussing the Ir moment size, we should determine where the implanted muon is located in Sr$_{1.9}$La$_{0.1}$IrO$_4$. The previous μSR study [29] stated that there are two main types of muon sites in Sr$_2$IrO$_4$; one near the apical oxygen, the other near the in-plane oxygen. However, the implanted muons do not occupy these sites equally; more than 90% of the muons are located in the apical oxygen site ($\theta = 60^\circ$, $\phi = 45^\circ$, $r = 1$ Å from the apical oxygen) of Sr$_2$IrO$_4$. We thus assume that all implanted muons occupy the apical oxygen site in...
Sr$_1$gLa$_{0.1}$IrO$_4$ (as in Sr$_2$IrO$_4$), because both crystal structures ($x = 0$ and $x = 0.1$) are nearly unchanged.

We now estimate the Ir moment size of Sr$_1$gLa$_{0.1}$IrO$_4$. In Sr$_2$IrO$_4$, the moment size of the $J_{\text{eff}} = 1/2$ pseudospin is reduced to $\sim 0.4 \, \mu_B$, as confirmed by previous $\mu$SR, RIXS, and neutron diffraction measurements [9,29,30]. Although a pseudospin of $J_{\text{eff}} = 1/2$ is almost equivalent to the superposition of a magnetic dipole and an octopole, the local field $B_{\text{loc}}$ at the muon site is given by the vector summation of the pointlike magnetic dipoles situated at the Ir sites, because the octopole makes a relatively small contribution to reducing the total magnetic induction. Note that the transferred moment from Ir to O is negligibly small (a few percent of $0.4 \, \mu_B$) in Sr$_2$IrO$_4$ [29]. The Gaussian linewidth $\Delta$ is given as the dipole summation,

$$\Delta = \left[ \frac{2}{3} \sum_{\mu \beta} \left( |\hat{A}_j \mu_j| \right)^2 \right]^{1/2},$$

for possible muon sites, where $\hat{A}_j$ is the dipole tensor expressed as

$$\hat{A}_j = \frac{1}{r_{ij}^3} \left( \frac{3\alpha \beta}{r_j^2} - \delta_{\alpha \beta} \right) \langle \alpha, \beta = x, y, z \rangle.$$

The summation runs through the $j$th Ir moments $\mu_j$ located at $r_j = \{x_j, y_j, z_j\}$ from a given muon site. The local field $B_{\text{loc}}$ at the muon site is given by

$$B_{\text{loc}} = \sum_j \hat{A}_j \mu_j.$$

Therefore, the root-mean-square value of the local field ($B_{\text{loc}}$)$_{\text{rms}}$ is proportional to the Gaussian linewidth $\Delta$. Each muon will experience magnetic fields from the local sites that obey the same Gaussian distribution.

As noted earlier, the analysis of the LF-$\mu$SR data yielded $\Delta = 4.47(3) \, \mu s^{-1}$ for Sr$_1$gLa$_{0.1}$IrO$_4$. By substituting this value into Eq. (5) and calculating the dipolar summation using the DIPELEC205 code [28], the effective Ir moment size $|\mu_{\text{eff}}|$ was determined as $0.0452(3) \, \mu_B$. This value is about one-tenth that of Sr$_2$IrO$_4$ ($0.4 \, \mu_B$) [9,29,30], so the decrease of the Ir moment is attributed to the effects of electron doping by La substitution. Incidentally, the spontaneous magnetization in the short-range ordered phase also reduces the value to less than one-tenth in the $x = 0.1$ sample [see Fig. 5(a)]. This suggests that the Ir moment is reduced even in the short-range ordered phase, consistent with the fact that such a small ordered moment ($0.045 \, \mu_B$) is difficult to resolve in neutron diffraction.

Although we have clarified the short-range magnetic order in Sr$_1$gLa$_{0.1}$IrO$_4$, how the long-range order changes to a short-range order with electron doping in the Sr$_2$IrO$_4$ system remains unclear. To clarify the magnetic phase diagram in the Sr$_2$IrO$_4$ system, we will perform $\mu$SR experiments in electron-doped Sr$_2$La$_{1-x}$IrO$_4$ ($x = 0.05 \, (T_N \sim 208 \, K), \ x = 0.075 \, (T_N \sim 198 \, K), \ x = 0.13 \, (T_N \sim 114 \, K)$) in future work.

**ACKNOWLEDGMENTS**

We would like to acknowledge K. Kawashima, S. Suzuki, and S. Yasuda for collaboration in the early stage of the experiment. We also would like to acknowledge M. Nagao and A. Miura for valuable discussions. The authors are grateful to K. Tomimoto at the Center for Instrumental Analysis for the scanning electron microscope measurements. The muon experiment at the Materials and Life Science Experimental Facility of the J-PARC was performed under a user program (Proposal No. 2018A0254). This work was supported by Grants-in-Aid from the Ministry of Education, Culture, Sports, Science and Technology (MEXT) under Grants No. 2500003, No. 26247057, No. 16K17750, and No. 2704, and was partially supported by the Electric Power Development Company Limited (Dengenkaihatsu) and Program for Advancing Strategic International Networks to Accelerate for Circulation of Talented Researchers from the Japan Society for the Promotion of Science (R2705).
