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The size dependence of the magnetic phase transition in reentrant ferromagnet NiMn is investigated. Multilayer films of NiMn/Cu with thicknesses between 30 and 13 000 Å are prepared using the ion-beam sputtering method in an ultrahigh vacuum. The ferromagnetic (FM)–reentrant-spin-glass (RSG) transition temperature T_{RSG} is determined based on the irreversibility in the temperature-dependent dc-susceptibility $\chi(T)$. The Curie temperature T_C is assigned to an inflection point in the $\chi(T)$ curve. At thicknesses greater than 200 Å, the transition temperatures are analyzed based on finite-size scaling, and the shift parameter λ and the characteristic length D^0, at which the magnetic phase transition disappears, are evaluated. For the FM-RSG transition, $\lambda_{\mathrm{RSG}}=0.66\pm0.44$ and $D^0_{\mathrm{RSG}}\sim8$ Å are obtained. For the paramagnetic (PM)-FM transition, $\lambda_{\mathrm{FM}}=1.31\pm0.28$ and a remarkably large value of the characteristic length D^0_{FM}, 43 ± 13 Å, are evaluated. This large value of D^0_{FM} is discussed in connection with the inhomogeneous spin fluctuation appearing in the FM phase, which is found based on previous Mössbauer observation. The PM-FM and FM-RSG transition temperatures, as functions of thickness, intersect at a critical thickness D_c at which the FM phase disappears. Below D_c, in addition, the spontaneous magnetization disappears in the low-temperature phase. This observation indicates that there is a vertical boundary line through the thickness of D_c, which separates the reentrant ferromagnet, having a low-temperature RSG phase with ferromagnetic correlation, from the pure spin glass. This is compared with the magnetic phase diagrams of reentrant ferromagnetic systems.

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I. INTRODUCTION

In a reentrant ferromagnet, having a large number of antiferromagnetic couplings in addition to a majority of ferromagnetic couplings between the individual spins, the reentrant-spin-glass (RSG) phase appears at temperatures below the ferromagnetic (FM) phase. The magnetism of the RSG phase has been explained based on the two kinds of pictures, i.e., a mean-field-type picture[1,2] and an inhomogeneous picture explained based on the random-field effect.[3] In the former, the long-range FM order coexists with the spin-glass (SG) order in the RSG phase. In contrast, the latter predicts that the long-range ferromagnetic order is broken when the system undergoes a transition to the RSG phase. In addition, the FM phase, located at temperatures above the RSG phase, shows a dynamic behavior characterized by a chaotic nature similar to that of the spin-glass phase.[4] This characteristic, different from the robust nature of the regular ferromagnetic phase, has not been sufficiently interpreted based on the established models. Thus there remains some ambiguities related to the magnetism in the RSG and FM phases in the reentrant ferromagnet.

It is well known that the magnetic phase transition temperature in a magnetic thin film decreases as the film thickness D decreases. In a comparable thicker film where the magnetic phase transition temperature $T(D)$ deviates slightly from the bulk transition temperature T_b, the following expression, based on finite-size scaling, can be applied to analyze the thickness- dependent data:[5]

$$\frac{T_b-T(D)}{T_b}=(D/D^0)^{-\lambda}, \tag{1}$$

where D^0 is the characteristic length at which the magnetic phase transition disappears, and λ is the shift exponent which characterizes the sensitivity of the magnetic phase transition temperature to the decrease in the film thickness. In a film of conventional spin-glass material, e.g., CuMn and AgMn, the relation $\lambda_{\mathrm{SG}}<1$, theoretically predicted for the transition from the paramagnetic (PM) phase to the SG phase, was confirmed experimentally.[6–9] For the PM-FM transition temperature T_C in regular ferromagnetic films, in addition, a value λ_{FM} of 1–2 has been observed.[10–13] The FM-RSG transition temperature T_{RSG}, of a multilayer film containing a reentrant ferromagnet NiMn, was studied in a NiMn layer thickness ranging from 100 to 5000 Å,[14] and a scarce size dependence was observed. Due to the insufficient information regarding the size-dependent magnetic properties of the reentrant ferromagnet, however, there remain the following questions to be answered.

(1) How is the FM-RSG transition temperature dependent on film thickness in the thinner thickness region? Is it similar to that of the PM-SG transition in canonical spin glasses?

(2) Do differences exist in the size-dependent PM-FM transitions between reentrant ferromagnet and regular ferromagnetic materials? Does the chaotic nature of the FM phase in the reentrant ferromagnet reflect size dependence?

(3) Is it true that, provided the value of λ_{FM} for the PM-FM transition is larger than λ_{RSG} for the FM-RSG tran-

sition, as expected from previous studies of canonical spin glasses and regular ferromagnetic materials, only the SG phase can survive in the thinner thickness region? If this situation is realized, what is the magnetic nature of the low-temperature SG phase?

In the present study, we examine the multilayer film of the typical reentrant ferromagnet NiMn as a function of thickness in order to answer the above questions. The disordered alloy of NiMn with Mn concentrations from ~19% to 23.9% has the typical properties of a reentrant ferromagnet.[15] The RSG is phase has been characterized in terms of a mixed state of spin-glass ordering and ferromagnetism through the observation of a ferromagnetic domain using a transmission electron microscope,[16] and by means of a neutron depolarization analysis.[17] We prepared multilayer films of NiMn with a Mn concentration of ~22%, and determined the phase transition temperatures T_C and T_{RSG} in a thickness region of 30–13 000Å. The following characteristics were found in the size-dependent phase transitions: both values of λ_{RSG} and D^0_{RSG} for the FM-RSG transition are comparable with those of the canonical spin glasses. On the other hand, the magnitude of λ_{FM} for the PM-FM transition is intrinsically the same as that of regular ferromagnetic material, although the characteristic length D^0_{FM} is significantly large. As a result, the PM-FM transition is more sensitive to change in the film thickness compared with the FM-RSG transition. This results in the disappearance of the FM phase at thicknesses thinner than the critical thickness D_c (~60 Å). This kind of size-dependent instability can be related to the spatially inhomogeneous nature of spin dynamics inherent in the random magnetic system. In the same thickness region, in addition, a collapse of the spontaneous magnetization is observed in the lower-temperature phase. This correlation in magnetism in both magnetic phases is discussed in comparison with the magnetic phase diagram of a reentrant ferromagnet, in which a vertical phase boundary exists near the multicritical point, so as to separate the reentrant ferromagnet from the pure spin glass.[18]

II. SAMPLE PREPARATION AND CHARACTERIZATION

The sample was prepared on a quartz substrate at room temperature by a multitarget ion-beam sputtering system in an ultrahigh-vacuum chamber (base pressure $\sim 10^{-9}$ Torr). The argon sputtering pressure was less than 4×10^{-4} Torr. The deposition rates were 0.22 and 0.38 Å/sec for NiMn and Cu, respectively. The thickness of the NiMn film ranged from 30 to 13 000 Å. For NiMn layer thicknesses below 1300 Å, the multilayer sample was prepared to obtain a magnetic signal sufficient for the magnetic measurement, in which a 300-Å Cu spacer was used to prevent the exchange coupling between NiMn layers.[14,19] A Cu capping layer of 600 Å was deposited on the multilayer to prevent the NiMn layer from oxidization. The periodicity in the NiMn/Cu multilayer was determined by analyzing the small-angle x-ray-diffraction data[20] with a Cu-$K\alpha$ line. The depth profile of the NiMn/Cu multilayer sample was also analyzed using secondary-ion-mass spectrometry (SIMS) in the area of 140 μm^2. The measurement was performed using an O_2^+ impact energy of 500 eV at an incident angle of 0°, i.e., normal to the film surface. The composition of the NiMn film was determined by means of inductively coupled plasma mass spectrometry (ICP MS).

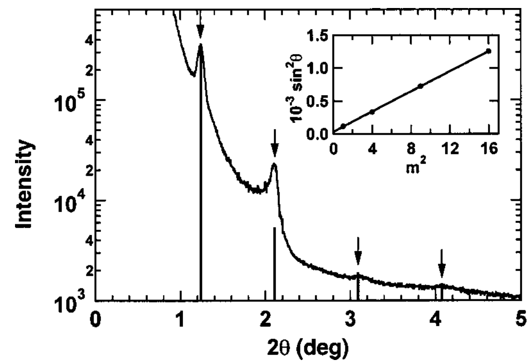

FIG. 1. Small-angle x-ray-diffraction data of a $[\mathrm{NiMn}52\,\text{Å}/\mathrm{Cu}36\,\text{Å}]_{30}$ multilayer sample. Four peaks (denoted by arrows), corresponding to the multilayer periodicity Λ, are observed. The solid lines indicate the best fit of the intensity to Eq. (3) with the parameter $d=12$ Å. The inset shows the $\sin^2\theta_m - m^2$ plot for the peaks, and the best fits to Eq. (2) and denoted by the solid line.

Figure 1 shows the x-ray-diffraction pattern of $[\mathrm{NiMn}52\,\text{Å}/\mathrm{Cu}36\,\text{Å}]_{30}$ in which four peaks, corresponding to the multilayer periodicity. Λ, are observed. The Bragg's law for the mth peak, in the small-angle region, is expressed as[21]

$$\sin^2\theta_m = \left(\frac{m\lambda_x}{2\Lambda}\right)^2 + 2\delta, \tag{2}$$

where θ_m is the peak angle, λ_x is the wavelength of the x ray, Λ is the multilayer periodicity, and 2δ is the compensation term for refraction. A linear relation between $\sin^2\theta_m$ and m^2 was observed (the inset in Fig. 1). This results in the periodicity Λ of 88 Å based on Eq. (2). Furthermore, the well-controlled periodicity of the multilayer film was also confirmed based on the depth profile of SIMS for $[\mathrm{NiMn}39\,\text{Å}/\mathrm{Cu}30\,\text{Å}]_{30}$ in which 30 peaks, related to NiMn and Cu layers, appeared alternately (Fig. 2). This level of quality of structure was maintained even for the thinnest NiMn layer. The width of the intermixing layer on the NiMn/Cu boundary was calculated based on the trapezoidal model, which is based on a kinematical diffraction theory.[22,23] The total scattering intensity I for the mth peak is expressed as

$$I = I_0 I_e |F(m)|^2 L(m), \tag{3}$$

where I_0 is the scale constant, I_e is the Lorentz polarization factor, $F(m)$ is the layer structure factor of the multilayer, which is inversely proportional to the width of the boundary intermixing layer d, and $L(m)$ is the Laue function which has a magnitude of N^2 (N is the number of the NiMn/Cu set) when the Laue condition is satisfied. We evaluated the interface width d of 12 Å using the least-squares fit of the decrement of the relative intensity of the four peaks to Eq. (3) (shown using the solid lines in Fig. 1). This corresponds to

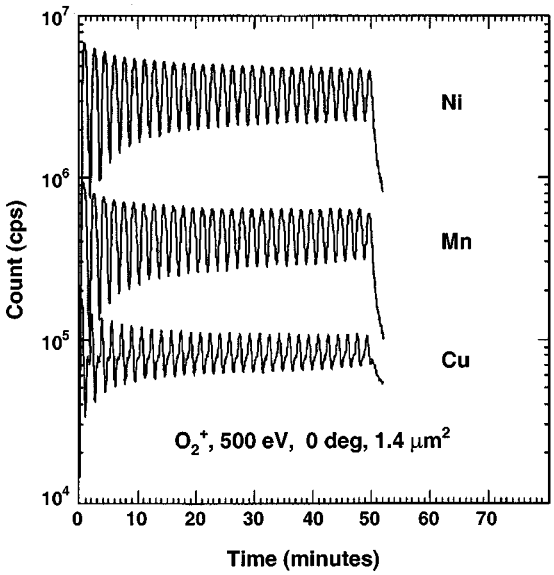

FIG. 2. SIMS depth profile of the $[\mathrm{NiMn}39\,\text{Å}/\mathrm{Cu}30\,\text{Å}]_{30}$ sample. Thirty alternating peaks corresponding to NiMn and Cu layers are observed.

three or four times the atomic spacing of the boundary intermixing layer if the bulk lattice constant is unchanged in the film.[24] The Mn concentration of 22 at. %, located in the reentrant ferromagnet regime, was determined from the ICP MS.

III. MAGNETIC DATA AND DISCUSSION

A. Thickness dependence of T_{RSG} and T_C

Magnetic measurements were performed using a superconducting quantum interference device magnetometer. The temperature dependent DC susceptibility χ, defined as the ratio of the magnetization M to the applied field H, was obtained under zero-field-cooled (ZFC) and the field-cooled (FC) conditions in order to evaluate the magnetic phase transition temperatures, i.e., T_{RSG} and T_C. In order to evaluate the spontaneous magnetization in the RSG phase, magnetization was measured at 6 K as a function of the field H_{cool} which was applied parallel to the film surface during cooling the sample from a temperature above T_C.[15]

Figures 3(a) and 3(b) show the temperature dependence of the dc susceptibility of NiMn thin films, with thicknesses ranging from 30 to 13 000 Å. We can observe the thermal irreversible behavior between ZFC and FC data at lower temperatures in all the samples. At thicknesses greater than 91 Å, in addition, the plateau region can be observed around 40 K, although it becomes narrow as the thickness decreases. These characteristics, observed above 91 Å, are typical of the reentrant ferromagnet. Below 65 Å, a broad peak replaces the plateau. This suggests the change in the magnetic nature of the NiMn film appearing between 65 and 91 Å, as mentioned below. Based on this temperature-dependent dc susceptibility, the magnetic transition temperature can be evaluated as functions of the film thickness. The spin freezing temperature T_{RSG} and the Curie temperature T_C are assigned

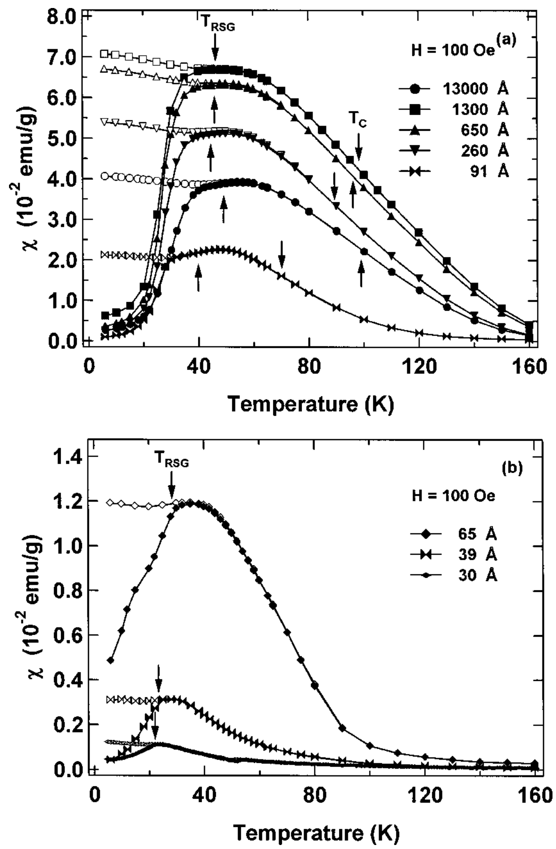

FIG. 3. Temperature dependence of the dc susceptibility χ in samples with thicknesses between 30 and 13 000 Å. The error in the dc susceptibility χ is smaller than the plot size.

to a temperature at which the irreversibility appears between FC and ZFC data and an inflection point[25] appearing at a temperature higher than the plateau region, respectively, where these are denoted by arrows in Fig. 3. We note that the spin freezing temperature of 49 K, evaluated in the thickest sample, corresponds to a Mn concentration of 22 at. % in the magnetic phase diagram,[15] which completely agrees with the result of ICP MS. On the other hand, the Curie temperature of 99 K in the thickest sample is significantly lower than that of a bulk sample having the same Mn concentration.[26] This may reflect the sensitivity of the FM phase to the change in sample preparation condition. Figure 4 shows the thickness dependences of T_{RSG} and T_C. First we pay attention to the behavior of T_{RSG}. As the thickness decreases from 13 000 to 130 Å, T_{RSG} shows a decrease of 8 K. This decrease is consistent with the previous result of a RSG-NiMn/Cu multilayer film.[14] In the thickness region thinner than 91 Å, T_{RSG} shows a steplike decrease. This singular change will be connected with the instability of the long-range FM order in Sec. III B. Provided that the data with $D > 200\,\text{Å}$ are analyzed using Eq. (1),[7] the best fit is realized using a characteristic length $D^0_{RSG} = 8^{+17}_{-8}\,\text{Å}$ and a shift exponent $\lambda_{RSG} = 0.66 \pm 0.44$ as denoted by the dotted curve in Fig. 4, where the comparative large error of the parameters is attributed to the low precision for a determination of T_{RSG}, based on the ZFC and FC curves of $\chi(T)$. The present value of D^0_{RSG} is comparable to the value reported for the other typical spin glasses, as shown in Table I. The shift exponent is intrinsi-

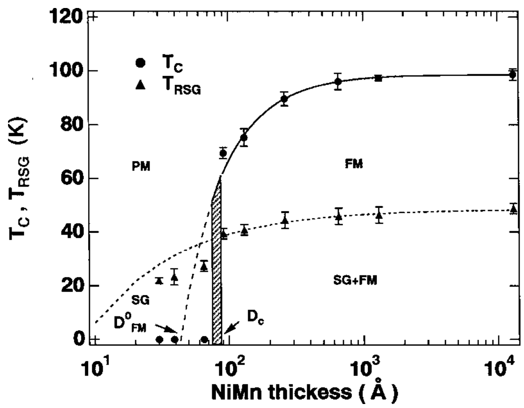

FIG. 4. Thickness dependence of T_{RSG} (denoted by the open triangle) and T_C (denoted by the open circle). The dotted and solid curves indicate the best fits of T_{RSG} and T_C to Eq. (1), respectively.

cally the same as that evaluated for the other spin glasses[6–9,27] and in a Monte Carlo simulation of Ising spin glass.[28] After all, the size dependence of T_{RSG} is indistinguishable from that of canonical spin glasses, except for the behavior at thicknesses thinner than 91 Å.

Next we focus on the Curie temperature T_C. The decrease in T_C is more remarkable compared with T_{RSG}, e.g., a decrease of 23 K, observed as the thickness decreases from 13 000 to 130Å, is three times larger than that of T_{RSG}. In the range with $D > 200$ Å, T_C was analyzed as a function of thickness based on finite-size scaling. The best fit is observed using a characteristic length D^0_{FM} of 43 ± 13 Å and a shift exponent λ_{FM} of 1.31 ± 0.28. The value of D^0_{FM} is significantly larger than that of the ferromagnetic Ni, which is on the order of several monolayers.[10,11] The shift exponent λ_{FM} is almost the same as that of ferromagnetic Ni, as shown in Table I.[10,11] The large value of D^0_{FM} implies that the ferromagnetic ordering in a reentrant ferromagnet is easily broken as the thickness decreases, in comparison with the regular ferromagnetic materials. This instability can be related to the Mössbauer observation in the FM phase of a NiMn reentrant ferromagnet,[29] i.e., small regions consisting of spins with large amplitudes of fluctuation existing in the ferromagnetic matrix. Provided that the film thickness is comparable to the characteristic length scale of this region, the long-range ferromagnetic order should be unstable. Thus the present evaluation of D^0_{FM} can predict the size of the spin fluctuation region (~40 Å). Thus this kind of spin fluctuation can bring about a distinction of size-dependent behavior between the FM phase in a reentrant ferromagnet and the regular ferromagnetic phase. In addition, this may be related to the chaotic behavior observed in the FM phase.[4] On the other hand, the agreement of λ_{FM} between NiMn and Ni suggests that the temperature-dependent change in the ferromagnetic correlation intrinsically originates from the ferromagnetic Ni-Ni pair correlation. At the thinner thickness region, the determination of T_C becomes ambiguous because the plateau region, observed in the temperature-dependent dc susceptibility, becomes indistinct. At thicknesses below 65 Å, we cannot observe any characteristic peculiar to the FM phase. In addition, we should note that the crossover of the thickness-dependent curves of T_{RSG} and T_C appears at approximately 60 Å (Fig. 4). This coincidence is consistent with an expectation based on the difference of parameters D^0 and λ between two phase transitions, i.e., only one magnetic phase appearing below the PM phase at thicknesses thinner than the critical thickness D_c. Thus we claim that the FM phase disappears in the NiMn thin film below $D_c \sim 60$ Å. This will be supported in Sec. II B by an evaluation of the spontaneous magnetization at a low temperature.

TABLE I. The shift exponent λ and the characteristic length D^0 for spin glass and ferromagnetic materials. MC shows the Monte Carlo simulation.

	Material	λ	D^0 (Å) or (ML)
Spin glass	NiMn[a]	0.66 ± 0.44	8^{+17}_{-8}
	CuMn/Si[b]	0.63 ± 0.15	30~35
	CuMn/Cu[c]	0.91 ± 0.25	0~10
	$CuMn/Al_2O_3$[d]	0.64 ± 0.07	20 ± 1
	AuFe[e]	-	100
	AgMn[f]	0.77 ± 0.12	-
	MC of Ising system[g]	0.77	-
Ferromagnet	NiMn[a]	1.31 ± 0.28	43 ± 13
	Ni(poly)[h]	1.33 ± 0.13	6 ± 0.3 ML
	Ni(111)[i]	1.4	2ML

[a]Present study.
[b]Reference 6.
[c]Reference 7.
[d]Reference 8.
[e]Reference 27.
[f]Reference 9.
[g]Reference 28.
[h]Reference 10.
[i]Reference 11.

B. Magnetic phase diagram of reentrant ferromagnet NiMn thin film

In this section, we propose a magnetic phase diagram of a reentrant ferromagnet NiMn in the form of a thin film. For this purpose, the magnetic nature of the low-temperature phase should be clarified at thicknesses thinner than D_c. The thermal irreversible behavior, observed in the ZFC and FC susceptibilities, is observed below the cusp temperature in a thickness regime below 65 Å [Fig. 3(b)]. In addition, a unidirectional anisotropy, peculiar to the spin glass NiMn, can be observed down to the thinnest thickness (30 Å), although the magnetization curve will be shown in a following paper.[30] Thus a spin-glass nature is retained even in the thinnest sample in the present work. Next we examine the spontaneous magnetization in the low-temperature phase according to the method of Abdul-Razzaq and Kouvel.[15] The magnetization, which was measured at a field of H_{cool} after field cooling the sample from 300 K to the measurement temperature at the same field, was plotted as a function of the applied field in Fig. 5. From an extrapolation in the low-field range to $H_{cool} = 0$ with a straight line, the spontaneous mag-

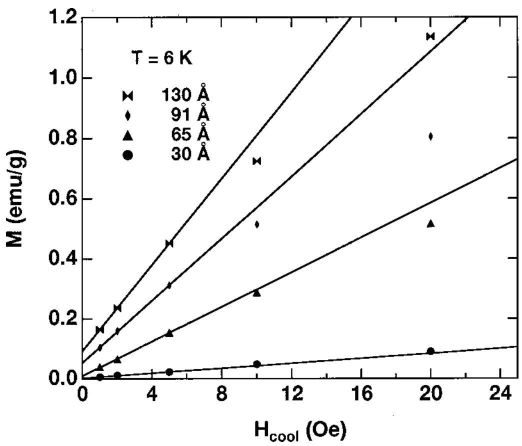

FIG. 5. A magnetization at 6 K is plotted as a function of H_{cool} for several samples with various thicknesses. The solid line indicates the least-square fits in the low H_{cool} range.

netization can be verified at 130 and 91 Å. In thinner samples of 65 and 30 Å, a zero spontaneous magnetization is observed. Thus the magnetism of the low-temperature phase, in a sample with a thickness below 65 Å, can be characterized by a pure spin-glass nature without long-range ferromagnetic correlation. This can explain the steplike change in the spin freezing temperature appearing between 91 and 65 Å, because the change from a RSG-FM phase transition to a SG-PM phase transition should bring about a singular change in the transition temperature as a function of film thickness. In addition, we emphasize the following important perspective: the disappearance of the FM phase is accompanied by a simultaneous disappearance of the ferromagnetic correlation in the low-temperature phase. Therefore, we can propose the magnetic phase diagram of reentrant ferromagnet NiMn thin film as follows (Fig. 4).

(1) In the thickness region above D_c, which is located between 65 and 90 Å, as shown by the hatched area in Fig. 4, a conventional reentrant phase transition is observed; i.e., as the temperature is lowered, the system undergoes a transition from a PM phase to a FM phase, and further undergoes a transition to a RSG phase, in which the spin glass and long-range ferromagnetic correlation coexist.

(2) Below D_c, a pure spin-glass transition is observed, i.e., as the temperature is lowered, the system undergoes a transition from a PM phase to a SG phase, in which the long-range ferromagnetic correlation disappears.

It is interesting to note that this feature resembles the magnetic phase diagram of a reentrant ferromagnet, proposed based on the mean-field picture, in which a low-temperature RSG phase with ferromagnetic correlation is separated from a pure spin glass phase by a vertical boundary line through a multicritical point.[18] In addition, a similar vertical boundary line, between RSG and SG phases, was also observed in the magnetic phase diagram of a reentrant antiferromagnetic system $Fe_xMn_{1-x}TiO_3$.[31] This suggests that the relation between the disappearance of a FM phase and the successive disappearance of the ferromagnetic ordering in a spin-glass phase is a universal characteristic in the reentrant ferromagnetic system. Therefore, we claim that the study of the size-dependent magnetic behavior of a reentrant ferromagnet is very effective in obtaining direct evidence of this relation.

IV. CONCLUSION

The size dependence of the magnetic phase transition temperature in the reentrant ferromagnet NiMn was investigated using well-controlled multilayer films of NiMn/Cu. At thicknesses greater than 200 Å, the PM-FM and FM-RSG transition temperatures were analyzed based on finite-size scaling. The remarkably large value of the characteristic length D^0_{FM}, observed for the PM-FM transition, indicated the spatially inhomogeneous spin dynamics appearing in the FM phase which was found based on the previous Mössbauer observation. The two curves of the thickness-dependent transition temperatures, obtained using the best-fit parameters of D^0 and λ, intersect at a critical thickness D_c. The disappearance of the ferromagnetic characteristic was observed below D_c. In the same thickness region, in addition, the spontaneous magnetization disappears in the low-temperature phase. After all, the ferromagnetic ordering is simultaneously broken in FM and RSG phases when the thickness decreases to D_c, Thus there is a vertical boundary line through the thickness of D_c, which separates the reentrant ferromagnet, having a low-temperature RSG phase with a ferromagnetic correlation, from the pure spin glass in the NiMn system. This observation is a common characteristic of magnetic phase diagrams of reentrant ferromagnetic systems.

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[26] The Curie temperature of reentrant ferromagnet has been generally determined as a temperature at which the dc susceptibility $\chi(T)$ shows a sharp kink due to a deviation from the demagnetization-limited value in a sufficiently weak applied field. As a higher field is applied, this method is invalid due to the disappearance of the plateau region. As mentioned in Ref. 25, the inflection point in $\chi(T)$, intrinsically independent of the amplitude of the applied field, is an index of the Curie temperature as well as the sharp kink in low-field susceptibility. In the form of a thin film, the demagnetization-limit cannot be observed under a field applied parallel to the film surface. Thus the inflection point is used to determine the Curie temperature in the present work. In addition, we note that the inflection point temperature in $\chi(T)$ of the 1300-Å sample, evaluated in a field of 20 Oe, is the same as that for 100 Oe within experimental error.
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