Observation of a magnetic-field-induced Griffiths phase in three-dimensional Ising random magnet $Ni_pMg_{1-p}(OH)_2$ from absorption of AC magnetic susceptibility

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(Dated: October 15, 2009)

The nature of the Griffiths phase in three-dimensional Ising random magnet $Ni_pMg_{1-p}(OH)_2$ (p = 0.10, 0.25, 0.315, 0.50, 0.80, and 1) is studied from the measurements of absorption χ'' (the out-of phase in AC magnetic susceptibility) in the field-cooled (FC) state. The temperature dependence of χ'' is measured in the vicinity of the critical temperature [Néel temperature for $p = 0.50$, 0.80, 1, and the spin glass transition temperature T_{SG} for $p = 0.1$, 0.25, 0.315] in the presence of an external magnetic field H. For $p = 0.50$, 0.80, and 1, a peak of χ'' vs T due to the metamagnetic transition drastically shifts to the low temperature side with increasing H, while a broad peak of χ'' vs T is clearly observed well above the Néel temperature $T_N(H = 0)$ for $H > 20$ kOe. This result gives a piece of evidence for the magnetic-field induced Griffiths phase, where the Griffiths temperature T_G is defined as the peak temperature of χ'' vs T above $T_N(H = 0)$. This T_G shifts to the low temperature side with increasing H, reflecting the nature of antiferromagnetic fluctuations. The peak height $\chi''(T_G)$ drastically increases with increasing H for $H > 20$ kOe, following a power form, $\chi''(T_G) \approx H^{m-1}$. The exponent m is 3.12 ± 0.04 for $p = 1$, 2.96 ± 0.03 for $p = 0.80$, and 2.47 ± 0.06 for $p = 0.50$. For $p = 0.10$, 0.25, and 0.315, a broad peak of χ'' vs T is clearly observed well above $T_{SG}(H = 0)$ for $H > 20$ kOe. The Griffiths temperature T_G shifts to the high temperature side with increasing H, reflecting the nature of ferromagnetic fluctuations. The peak height $\chi''(T_G)$ drastically increases with increasing H for $H > 20$ kOe, following a power form, $\chi''(T_G) \approx H^{m-1}$. The exponent m is 2.28 ± 0.08 for $p = 0.315$, 2.20 ± 0.07 for $p = 0.25$, and 2.35 ± 0.07 for $p = 0.10$.

PACS numbers: 75.50.Lk, 75.40.Gb, 75.30.Kz

I. INTRODUCTION

The observation of the Griffiths phase has been reported in site-diluted Ising systems with a concentration (p) of magnetic ions which has a percolation threshold p_c.[1–4] The Griffiths phase refers to a temperature region between a critical temperature $T_c(p)$ associated with the long range spin order and a so-called Griffiths temperature T_G [$> T_c(c)$].[5] The nature of the Griffiths phase is characterized by a statistically rare, but large clusters of unfrustrated spins over the whole system. Such clusters become quasi-ordered below T_G and flips collectively with anomalously long characteristic times. The consequence of such processes is slow dynamics of the spin autocorrelation functions $q(t)$ (= $[\langle S_i(0)S_i(t)\rangle]$), whose asymptotic form at large time t is given by an enhanced power law

$$q(t) \approx \exp[-A(\ln t)^{d/(d-1)}], \tag{1}$$

for the Ising systems with short-ranged interaction, where A is a constant, d is the spatial dimension and $\langle\cdots\rangle$ and $[\cdots]$ in the expression of $q(t)$ represent thermal and disorder averages respectively. The picture of this cluster dynamics can be natively extended to spin glasses by redefining a cluster as a group of spins connected with few frustrations.[5–8]

In this paper we report the observation of the Griffiths phase in a diluted Ising random magnet $Ni_pMg_{1-p}(OH)_2$ (the Ni concentration p)[9–16] using the absorption χ'' of the AC magnetic susceptibility. The sample in the present work was the same as that used in the previous papers (powdered samples).[14,15] We show that the Griffiths phase is strongly enhanced by the application of magnetic fields for $0.1 \le p \le 1$. The absorption χ'' vs T shows a broad peak centered at $T_G(H)$. The peak height of χ'' drastically increases with increasing H, forming the magnetic-field-induced Griffiths phase. For $0.50 \le p \le 1$ the system exhibits a metamagnetic transition at high magnetic field. The metamagnetic transition temperature $T_N(H)$ greatly decreases with increasing the magnetic field (typically $H > 20$ kOe), while the Griffiths temperature $T_G(H)$ slightly decreases with increasing H. For $0.10 \le p \le 0.315$, the metamagnetic behavior no longer exists. In turn a spin glass phase newly appears. There is a phase boundary between this spin glass phase (lower temperature phase) and the Griffiths phase (higher temperature phase). The Griffiths phase is strongly enhanced by the application of H. The Griffith temperature T_G slightly increases with increasing H.

II. BACKGROUND

In a $Ni(OH)_2$, Ni^{2+} ions form a triangular lattice with a lattice constant $a = 3.117\mathring{A}$ in the c plane. The separation distance between adjacent Ni^{2+} layers is $c = 4.595\mathring{A}$. The antiferromagnetic interplanar exchange interactions ($J_2 = -0.28$ K and $J_3 = -0.09$ K) are weaker than the ferromagnetic intraplanar exchange interaction ($J_1 = 2.7$ K). Because of a negative uniaxial single ion anisotropy D ($= -0.8$ K), magnetic moments of Ni^{2+} spin ($S = 1$) are directed along the c axis. $Ni(OH)_2$ undergoes an antiferromagnetic phase transition at the Néel temperature T_N ($= 26.5$ K). Below T_N the 2D ferromagnetic long range

order is established in each Ni^{2+} layer. Such 2D ferromagnetic layers are antiferromagnetically coupled along the c axis, forming a three-dimensional (3D) antiferromagnetic order. The numbers of neighbors coupled by the interactions J_1, J_2, and J_3, are denoted by z_1 (= 6), z_2 (= 2), and z_3 (= 12), respectively. The crystal structure and the definition of J_1, J_2, J_3 are given in our previous papers.[14,15]

$Ni_pMg_{1-p}(OH)_2$ is a diluted 3D Ising random magnet, where a part of Ni^{2+} ions are randomly replaced by nonmagnetic Mg^{2+} ions.[9–15] The Néel temperature T_N decreases with decreasing concentration p for $p > 0.4$. The percolation threshold p_c where the critical temperature reduces to zero, is about $p_c = 0.1$. The curve of T_N against p shows a terraced form in the concentration region below $p = 0.4$: T_N is extrapolated to zero near $p_1 = 0.33$. The value of $p_1 = 1/3$ coincides with the estimated critical concentration when only the first neighbor interaction is taken into account; $p_1 = 2/z_1 = 1/3$. This value of p_c (= 0.1) coincides with the estimated critical concentration when the first neighbor interaction is taken into account; $p_c = 2/(z_1 + z_2 + z_3) = 1/10$. As will be discussed in Sec. V,[15] the magnetic phase diagram of $Ni_pMg_{1-p}(OH)_2$ consists of spin glass (SG) phase and Griffiths phase for $0.1 \leq p \leq 0.315$, reentrant spin glass (RSG) phase, antiferromagnetic (AF) phase, and the Griffiths phase for $0.5 \leq p \leq 0.8$, and the AF phase and the Griffiths phase for $p = p_c = 0.1$.

III. EXPERIMENTAL PROCEDURE

A. ZFC and FC magnetic susceptibility at $H = 1$ Oe

Before setting up a sample at 298K, a remnant magnetic field in a superconducting magnet was reduced to one less than 3 mOe using an ultra low field capability option of the SQUID (superconducting quantum interference device) magnetometer (MPMS XL-5, Quantum Design). For convenience, hereafter this remnant field is denoted as the state of $H = 0$. The sample was cooled from 298 to 1.9 K at $H = 0$. Then an external magnetic field H (= 1 Oe) was applied at 1.9 K. A zero-field cooled magnetization (M_{ZFC}) was measured with increasing T from 1.9 to 50 K. After the ZFC measurement, the sample was heated and kept at 100 K for 20 min. It was again cooled to 30 K in the presence of the same H. A field-cooled magnetization (M_{FC}) was measured with decreasing T from 30 to 1.9 K. The ZFC and FC magnetic susceptibilities are defined as $\chi_{ZFC} = M_{ZFC}/H$ and $\chi_{FC} = M_{FC}/H$.

B. Measurement of the AC magnetic susceptibility in the FC state

The T dependence of the dispersion χ' and the absorption χ'' was measured in the presence of various magnetic field H, where the amplitude of AC magnetic field h is typically 2 or 3 Oe and the frequency is $f = 1$ Hz. The sample was cooled from 298 to 1.9K at $H = 0$. An external magnetic field ($0 < H < 50$ kOe) was applied at 1.9K. Then χ' and χ'' were simultaneously measured with increasing temperature T from 1.9 to 30 K in the presence of fixed H. After each T scan, H was changed at 30K and T was decreased from 30 to 1.9 K. Then the measurement was repeated at this H. This means that the AC susceptibility was measured in the FC state.

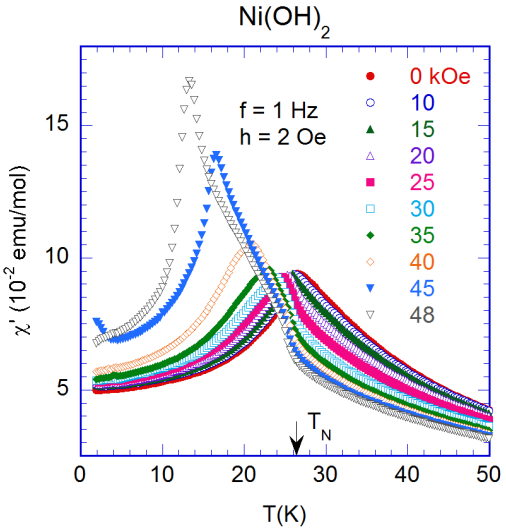

FIG. 1: (Color online) T dependence of χ' for $p = 1$ at various magnetic fields. H = 0, 10, 15, 20, 25, 30, 35, 40, 45, and 48 kOe. $T_N(H = 0) = 26.5$ K (denoted by arrow). $f = 1$ Hz. $h = 2$ Oe. Each measurement of χ' vs T was carried out in the FC state.

IV. RESULT

A. Magnetic-field-induced Griffiths phase in $p = 1$

In our previous paper[15] we have reported the temperature dependence of the ZFC and FC susceptibilities χ_{ZFC} and χ_{FC} for $Ni(OH)_2$ in the presence of $H = 1$ Oe. It is found that the difference δ ($= \chi_{FC} - \chi_{ZFC}$), appears below a characteristic temperature, and increases with decreasing T. Here we define this temperature as the Néel temperature T_N (= 26.5 K). This system shows a metamagnetic behavior in the presence of an external magnetic field. The critical field for the metamagnetic transition is $H_c = 55 \pm 1$ kOe at $T = 1.56$ K (Enoki et al.[12]).

Figure 1 shows the T dependence of the dispersion χ' in the FC state for $p = 1$ at various magnetic fields. The dispersion shows a sharp peak at T_N at $H = 0$. No anomaly associated with the Griffiths phase is clearly observed near T_N. The shift of the peak of χ' to the low temperature side with increasing H is due to the meta-

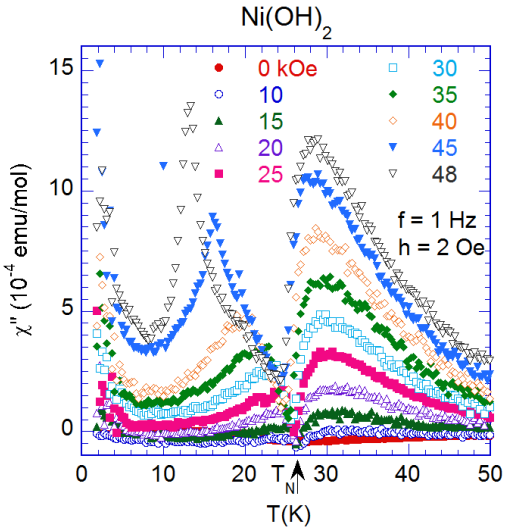

FIG. 2: (Color online) T dependence of χ'' for $p = 1$ at various magnetic fields. $H = 0$, 10, 15, 20, 25, 30, 35, 40, 45, and 48 kOe. $f = 1$ Hz. $h = 2$ Oe. Each measurement of χ'' vs T was carried out in the FC state. $T_N(H = 0) = 26.5$ K (denoted by arrow). Negative value of χ'' arises from the experimental uncertainty.

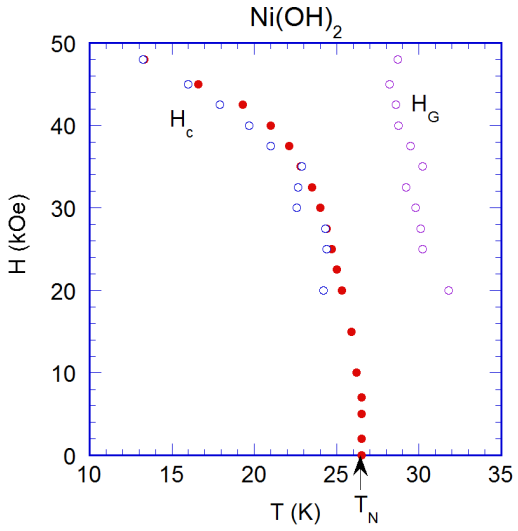

FIG. 3: (Color online) Magnetic H-T phase diagram of $Ni(OH)_2$: peak temperatures of χ' (closed circles) and χ'' (open circles). The H_G line denotes the phase boundary between the magnetic-field-induced Griffiths phase and the PM phase. The H_c line denotes the phase boundary between the AF phase and the magnetic-field-induced Griffiths phase. The critical field is $H_c = 55 \pm 1$ kOe at $T = 1.56$ K (Enoki et al.[12]). The Néel temperature $T_N(H = 0) = 26.5$ K (denoted by arrow).

magnetic transition between the AF phase and the PM phase (exactly the Griffiths phase). The metamagnetic transition temperature $T_N(H)$ drastically decreases with increasing H for $H > 35$ kOe.

Figure 2 shows the T dependence of the absorption χ'' for $p = 1$ at various magnetic fields, where $f = 1$ Hz. A peak due to the metamagnetic transition drastically shifts to the low temperature side with increasing H. No peak is observed around $T = T_N$ at $H = 0$. With increasing H, a broad peak of χ'' vs T starts to appear just above T_N. This peak slightly shifts to the low temperature side, while the peak height drastically increases with increasing H. This broad peak may be associated with the transition between the Griffiths phase and the PM phase. The separation between the peak due to the metamagnetic transition and the peak due to the appearance of the Griffiths phase becomes more prominent for $H > 35$ kOe.

Figure 3 shows the magnetic H-T diagram for $p = 1$. There are two lines, the H_G line and H_c line. The line H_G is the phase boundary between the Griffiths phase and the PM phase, while the line H_c is the phase boundary between the AF phase and the Griffiths phase. The critical field is $H_c = 55 \pm 1$ kOe at $T = 1.56$ K (Enoki et al.[12]).

Figure 4(a) shows the H dependence of $T_G(H)$ for $p = 1$. Here we define $T_G(H)$ as a peak temperature of χ'' vs T. This Griffiths temperature tends to decrease with increasing H, for example, $T_G(H = 20$ kOe$) = 31.8$ K and $T_G(H = 48$ kOe$) = 28.7$ K, reflecting the nature of the antiferromagnetic fluctuations. Note that $T_G(H)$ is relatively higher than the Néel temperature $T_N = 26.5$ K at $H = 0$ for $p = 1$. In Fig. 4(a) we also show the H dependence of the peak height $\chi''(T_G)$ for $p = 1$. This peak height is very small for $H < 20$ kOe. It drastically increases with increasing H above 20 kOe. The least squares fit of the $\chi''(T_G)$ vs H (4(b)) to

$$\ln \chi''(T_G) = A + \zeta \ln H, \tag{2}$$

for $20 < H < 48$ kOe, yields the values of A and ζ as

$$A = -14.291 \pm 0.15, \quad \zeta = 2.12 \pm 0.04,$$

where A and ζ are the fitted parameter, H is in the units of kOe, and $\chi''(T_G)$ is in the units of emu/mol. As will be introduced in Sec. V, the exponent m for the domain-wall magnetization M_{wall} is related to the exponent ζ as,

$$m = \zeta + 1 = 3.12 \pm 0.04. \tag{3}$$

B. Absorption χ'' at $p = 0.80$ in the presence of magnetic field

Figure 5 shows the T dependence of the absorption χ'' for $p = 0.80$ at various magnetic fields, where $f = 1$ Hz. A peak due to the metamagnetic transition drastically shifts to the low temperature side with increasing H. With increasing H, a broad peak of χ'' vs T associated with the Griffiths phase starts to appear well above

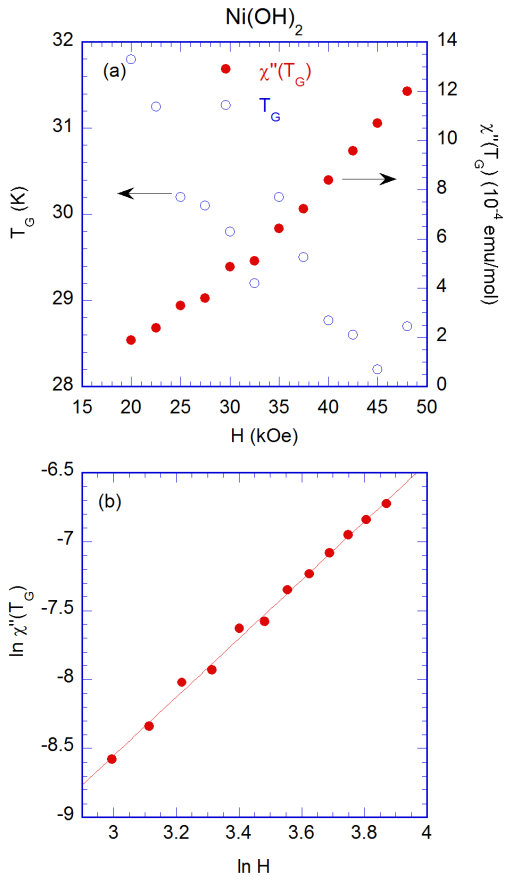

FIG. 4: (Color online) (a) Plot of the Griffiths temperature T_G as a function of the magnetic field H for $p = 1$. The Griffiths temperature T_G is defined as a temperature at which χ'' vs T has a peak for each H. T_G decreases with increasing H. Plot of the peak height $\chi''(T = T_G)$ as a function of H. The peak height drastically increases with increasing H for $H > 20$ kOe. (b) Plot of $\ln \chi''(T_G)$ vs $\ln H$, where H is in the units of kOe and $\chi''(T_G)$ is the units of emu/mol. The least-squares fitting curve is denoted by a straight line.

T_N. This peak slightly shifts to the low temperature side with increasing H, while the peak height drastically increases. The separation between the peak due to the metamagnetic transition and the peak due to the appearance of the Griffiths phase, becomes more prominent for $H > 20$ kOe. The peak due to the metamagnetic transition may disappear for $H > H_c$, where $H_c = 44 \pm 1$ kOe at $T = 1.56$ K (Enoki et al.[12]).

Figure 6(a) shows the H dependence of $T_G(H)$ for $p = 0.80$. This Griffiths temperature tends to decrease with increasing H, for example, $T_G(H = 20$ kOe$) = 24.78$ K and $T_G(H = 48$ kOe$) = 22.50$ K, reflecting the nature of the antiferromagnetic fluctuations (See Sec. V for detail). Note that $T_G(H)$ is relatively higher than the Néel temperature $T_N = 20.7$ K at $H = 0$ for $p = 0.80$. In Fig. 6(a) we also show the H dependence of the peak height $\chi''(T_G)$ for $p = 0.80$. The peak height drastically increases with increasing H above 20 kOe. The least

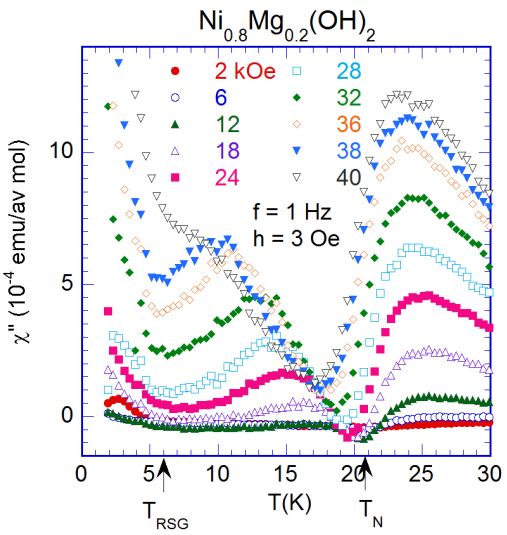

FIG. 5: (Color online) T dependence of χ'' for $p = 0.8$ at various magnetic fields. $H = 2$, 6, 12, 18, 24, 28, 32, 36, 38, and 40 kOe. The reentrant spin glass transition $T_{RSG} = 6$ K and the Néel temperature $T_N(H = 0) = 20.7$ K[14,15] (denoted by arrows). $H_c = 44 \pm 1$ kOe at $T = 1.56$ K (Enoki et al.[12]). $f = 1$ Hz. $h = 3$ Oe. Each measurement of χ'' vs T was carried out in the FC state.

squares fit of the plot $\ln \chi''(T_G)$ vs $\ln H$ (see Fig. 6(b)) to Eq.(2) for $18 \leq H \leq 48$ kOe, yields the values of A and ζ as

$$A = -13.93 \pm 0.11, \quad \zeta = 1.96 \pm 0.03.$$

The exponent m for M_{wall} is given by,

$$m = \zeta + 1 = 2.96 \pm 0.03.$$

C. Absorption χ'' at $p = 0.50$ in the presence of magnetic field

Figure 7 shows the T dependence of the absorption χ'' for $p = 0.50$ at various magnetic fields, where $f = 1$ Hz. A broad peak of χ'' vs T associated with the Griffiths phase starts to appear above T_N (= 10.1 K) for $H > 20$ kOe. The peak due to the metamagnetic transition may disappear for $H > H_c$, where $H_c = 20 \pm 5$ kOe at $T = 1.56$ K (Enoki et al.).

Figure 8(a) shows the H dependence of $T_G(H)$ for $p = 0.50$. This Griffiths temperature tends to decrease with increasing H, for example, $T_G(H = 20$ kOe$) = 18.07$ K and $T_G(H = 45$ kOe$) = 17.00$ K, reflecting the nature of the antiferromagnetic fluctuations. Note that $T_G(H)$ is relatively higher than the Néel temperature $T_N = 10.1$ K at $H = 0$ for $p = 0.50$. In Fig. 8(a) we also show the H dependence of the peak height $\chi''(T_G)$ for $p = 0.50$. This peak height drastically increases with increasing H above 20 kOe. The least squares fit of the plot $\ln \chi''(T_G)$

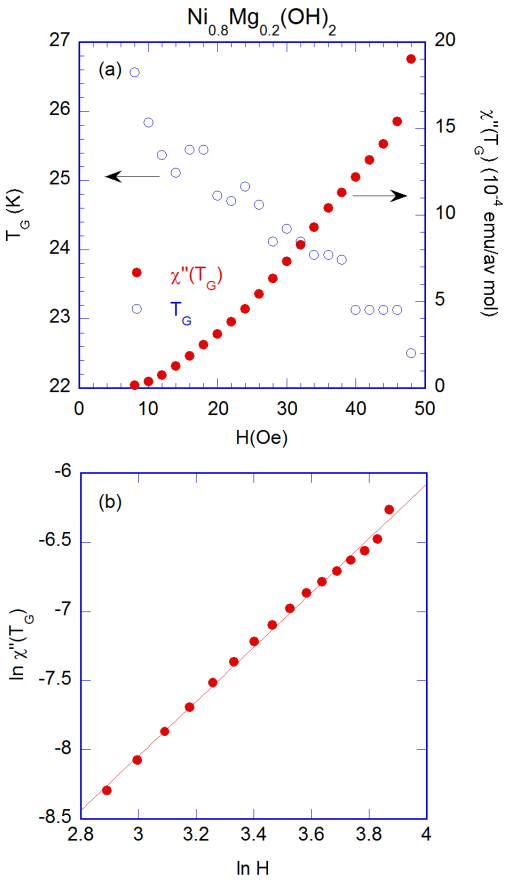

FIG. 6: (Color online) (a) Plot of T_G and the peak height $\chi''(T_G)$ as a function of H for $p = 0.80$. (b) Plot of $\ln \chi''(T_G)$ vs $\ln H$ for $18 \leq H \leq 48$ kOe, where H is in the units of kOe and $\chi''(T_G)$ is the units of emu/av mol. The least-squares fitting curve is denoted by a straight line.

vs $\ln H$ (see Fig. 8(b)) to Eq.(2) for $20 \leq H \leq 45$ kOe, yields the values of A and ζ as

$$A = -11.88 \pm 0.20, \quad \zeta = 1.47 \pm 0.06.$$

The exponent m for M_{wall} is given by

$$m = \zeta + 1 = 2.47 \pm 0.06.$$

D. Absorption χ'' at $p = 0.315$ in the presence of magnetic field

Figure 9 shows the T dependence of the absorption χ'' for $p = 0.315$ at various magnetic fields, where $f = 1$ Hz. A broad peak of χ'' vs T associated with the Griffiths phase starts to appear above the spin glass transition temperature T_{SG} (= 2.5 K), for $H > 20$ kOe.

Figure 10(a) shows the H dependence of $T_G(H)$ for $p = 0.315$. The Griffiths temperature tends to increase with increasing H, for example, $T_G(H = 20\ \text{kOe}) = 12.73$ K and $T_G(H = 40\ \text{kOe}) = 13.4$ K. Note that $T_G(H)$ is

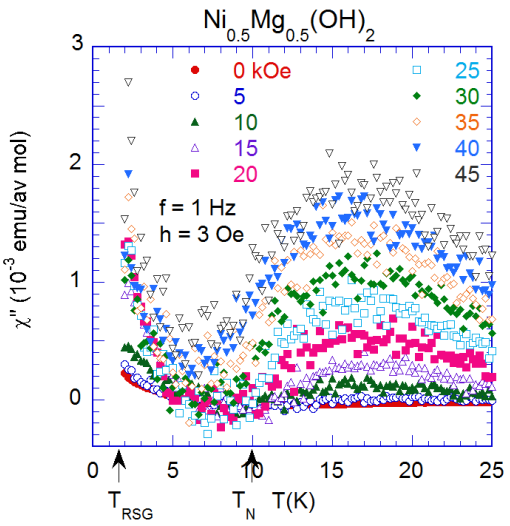

FIG. 7: (Color online) T dependence of χ'' for $p = 0.50$ at various magnetic fields. $H = 0$, 5, 10, 15, 20, 25, 30, 35, 40, and 45 kOe. $T_{RSG} = 5.3$ K and $T_N(H = 0) = 10.1$ K[14] (denoted by arrows). The critical field is $H_c = 20 \pm 5$ kOe at $T = 1.56$ K (Enoki et al.[12]). $f = 1$ Hz. $h = 3$ Oe. Each measurement of χ'' vs T was carried out in the FC state.

much higher than T_{SG} (= 2.5 K). This increase in $T_G(H)$ with H for $p = 0.315$ is in contrast with the decrease of $T_G(H)$ with H for $p = 0.50$. This increase of $T_G(H)$ may suggest the nature of the ferromagnetic fluctuations for $p = 0.315$. In Fig. 10(a) we also show the H dependence of the peak height $\chi''(T_G)$ for $p = 0.315$. This peak height drastically increases with increasing H above 20 kOe. The least squares fit of the plot $\ln \chi''(T_G)$ vs $\ln H$ (see Fig. 10(b)) to Eq.(2) for $20 \leq H \leq 40$ kOe, yields the values of A and ζ as

$$A = -11.27 \pm 0.25, \quad \zeta = 1.28 \pm 0.08.$$

The exponent m for M_{wall} is given by,

$$m = 2.28 \pm 0.08.$$

E. Absorption χ'' at $p = 0.25$ in the presence of magnetic field

Figure 11 shows the T dependence of the absorption χ'' for $p = 0.25$ at various magnetic fields, where $f = 1$ Hz. A broad peak of χ'' vs T associated with the Griffiths phase starts to appear above the spin glass transition temperature T_{SG} (= 2.5 K), for $H > 20$ kOe. Figure 12(a) shows the H dependence of $T_G(H)$ for $p = 0.25$. The increase of $T_G(H)$ increase with increasing H, for example, $T_G(H = 20\ \text{kOe}) = 12.63$ K and $T_G(H = 45$ kOe) = 13.97 K, reflects the nature of the ferromagnetic fluctuations. In Fig. 12(a) we also show the H dependence of the peak height $\chi''(T_G)$ for $p = 0.25$. This peak height drastically increases with increasing H above 20

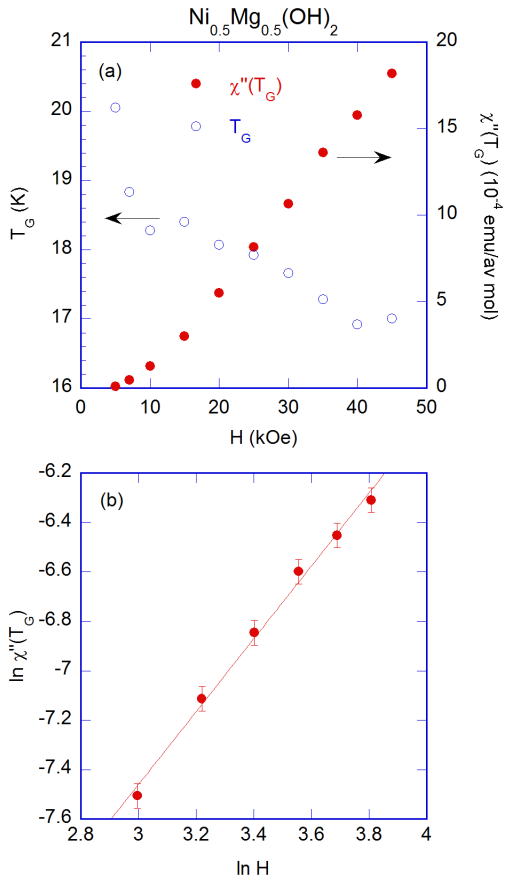

FIG. 8: (Color online) (a) Plot of T_G and the peak height $\chi''(T_G)$ as a function of H for $p = 0.50$. (b) Plot of $\ln \chi''(T_G)$ vs $\ln H$ for $20 \leq H \leq 45$ kOe, where H is in the units of kOe and $\chi''(T_G)$ is the units of emu/av mol. The least-squares fitting curve is denoted by a straight line.

kOe. The least squares fit of the plot $\ln \chi''(T_G)$ vs $\ln H$ (see Fig. 12(b)) to Eq.(2) for $20 < H < 45$ kOe, yields the values of A and ζ as

$$A = -11.21 \pm 0.24, \quad \zeta = 1.20 \pm 0.07.$$

The exponent m for M_{wall} is given by

$$m = 2.20 \pm 0.07.$$

F. Absorption χ'' at $p = 0.10$ in the presence of magnetic field

Figure 13 shows the T dependence of χ' for $p = 0.10$ at $H = 0$ was also measured at various f: $0.01 \leq f \leq 1000$ Hz, where $h = 0.5$ Oe. The dispersion χ' increases with decreasing T for $0.01 \leq f \leq 200$ Hz. It exhibits a broad peak around 2.05 K at $f = 330$ Hz, which slightly shifts to the high temperature side with increasing f. In contrast, χ'' starts to appear below 3 K and increases with decreasing T. The increase of χ'' with decreasing T below 2.1 K becomes more remarkable as f increases.

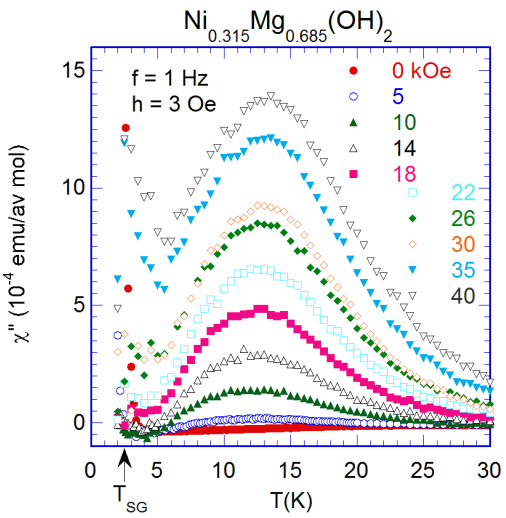

FIG. 9: (Color online) T dependence of χ'' for $p = 0.315$ at various magnetic fields. $H = 0$, 5, 10, 14, 18, 22, 26, 30, 35, and 40 kOe. $T_{SG} = 2.5$ K[14] (denoted by arrow). $f = 1$ Hz. $h = 3$ Oe. Each measurement of χ'' vs T was carried out in the FC state.

Figure 14 shows the T dependence of the absorption χ'' for $p = 0.10$ at various magnetic fields, where $f = 1$ Hz. A broad peak of χ'' vs T associated with the Griffiths phase starts to appear above the spin glass transition temperature T_{SG} (= 2.05 K), for $H > 20$ kOe. Figure 15(a) shows the H dependence of $T_G(H)$ for $p = 0.10$. The slight increase of $T_G(H)$ with increasing H, for example, $T_G(H = 20$ kOe$) = 13.3$ K and $T_G(H = 48$ kOe$)$ = 13.81 K, reflects the nature of the ferromagnetic fluctuations. In Fig. 15(a) we also show the H dependence of the peak height $\chi''(T_G)$ for $p = 0.10$. This peak height drastically increases with increasing H above 20 kOe. The least squares fit of the plot $\ln \chi''(T_G)$ vs $\ln H$ (see Fig. 15(b)) to Eq.(2) for $20 \leq H \leq 45$ kOe, yields the values of A and ζ as

$$A = -12.55 \pm 0.18, \quad \zeta = 1.35 \pm 0.05.$$

The exponent m for M_{wall} is given by

$$m = 2.35 \pm 0.07.$$

V. DISCUSSION

A. Domain wall magnetization

Here we discuss the physical meaning of the exponent m for the domain-wall magnetization M_{wall}.[17,18] The principal contribution at higher fields is due to the domain walls where parallel spins preferentially align with the field thus enhancing the magnetization. The bulk contribution is normally small and originates from a statistical imbalance between up and down spins within the

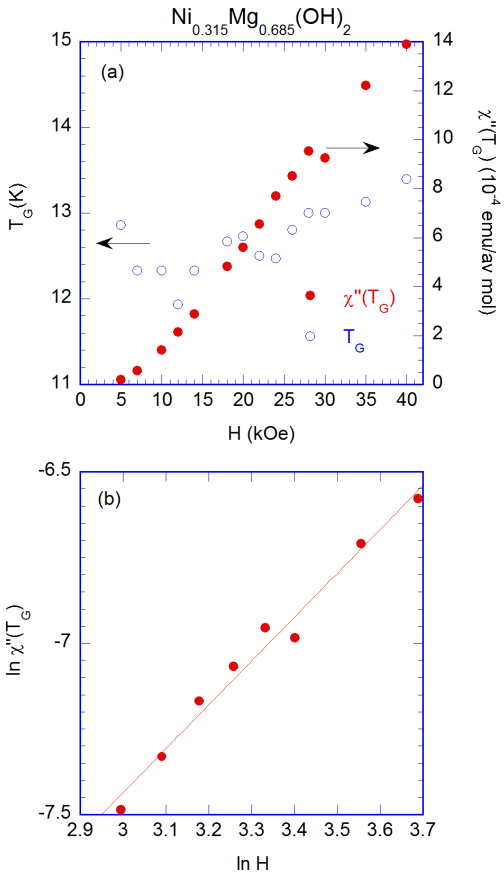

FIG. 10: (Color online) (a) Plot of T_G and the peak height χ'' $(T = T_G)$ as a function of H for $p = 0.315$. (b) Plot of $\ln \chi''(T_G)$ vs $\ln H$ for $20 \leq H \leq 40$ kOe, where H is in the units of kOe and $\chi''(T_G)$ is the units of emu/av mol. The least-squares fitting curve is denoted by a straight line.

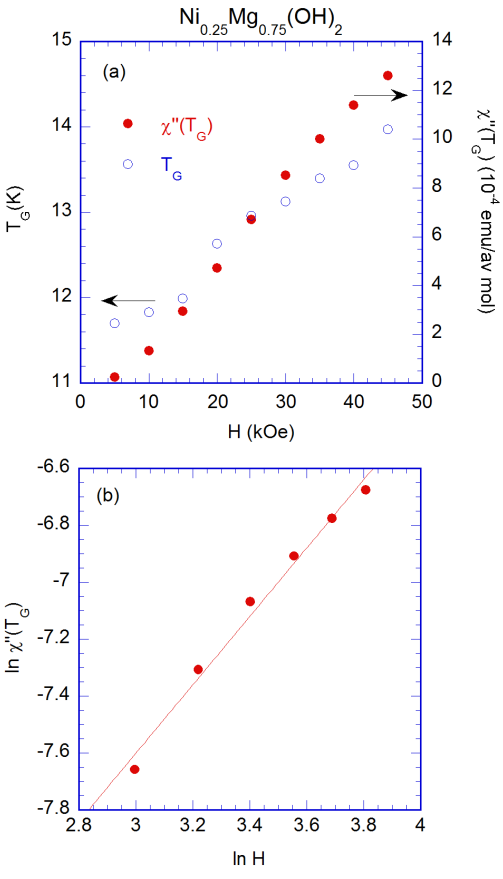

FIG. 12: (Color online) (a) Plot of T_G and the peak height χ'' $(T = T_G)$ as a function of H for $p = 0.25$. (b) Plot of $\ln \chi''(T_G)$ vs $\ln H$ for $20 \leq H \leq 5$ kOe, where H is in the units of kOe and $\chi''(T_G)$ is the units of emu/av mol. The least-squares fitting curve is denoted by a straight line.

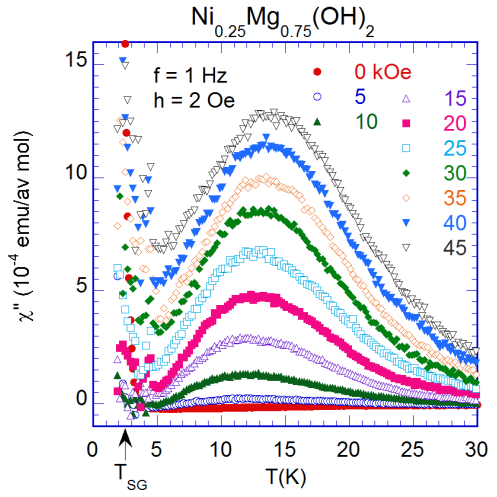

FIG. 11: (Color online) T dependence of χ'' for $p = 0.25$ at various magnetic fields. $H = 0$, 5, 10, 15, 20, 25, 30, 35, 40, and 45 kOe. $T_{SG} = 2.5$ K[14] (denoted by arrow). $f = 1$ Hz. $h = 2$ Oe. Each measurement of χ'' vs T was carried out in the FC state.

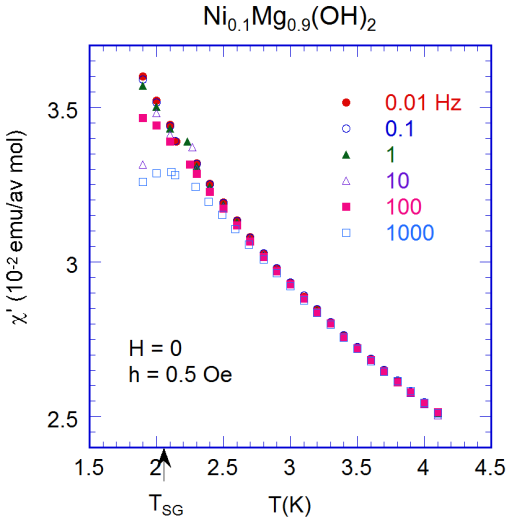

FIG. 13: (Color online) T dependence of χ' for $p = 0.10$ at various frequency. $H = 0$. $h = 0.5$ Oe.

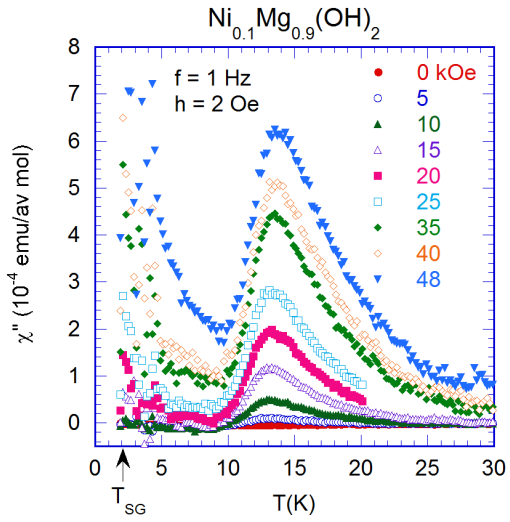

FIG. 14: (Color online) T dependence of χ'' for $p = 0.10$ at various magnetic fields. $H = 0$, 5, 10, 15, 20, 25, 30, 35, 40, and 48 kOe. $T_{SG} = 2.05$ K[14] (denoted by arrow). $f = 1$ Hz. $h = 2$ Oe. Each measurement of χ'' vs T was carried out in the FC state.

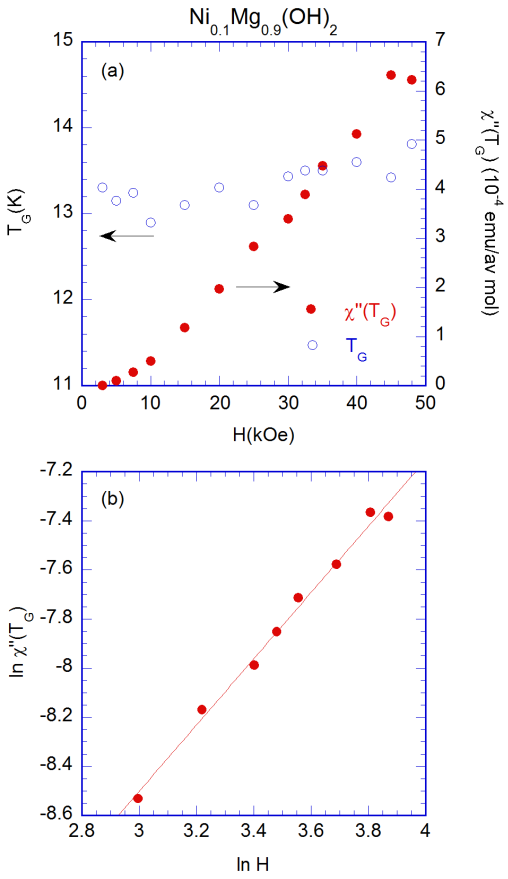

FIG. 15: (Color online) (a) Plot of T_G and the peak height χ'' $(T = T_G)$ as a function of H for $p = 0.10$. (b) Plot of $\ln \chi''(T_G)$ vs $\ln H$ for $20 \leq H \leq 45$ kOe, where H is in the units of kOe and $\chi''(T_G)$ is the units of emu/av mol.

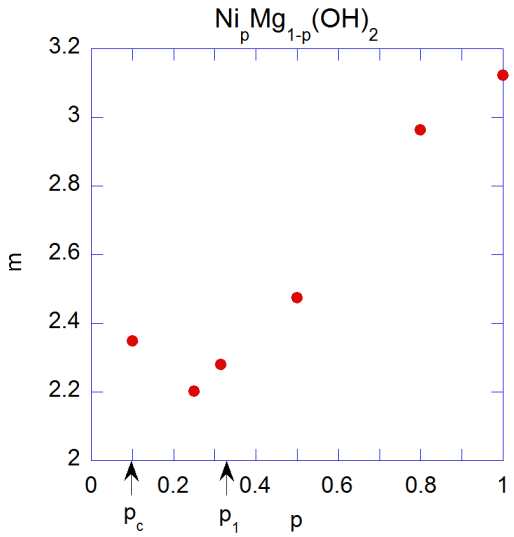

FIG. 16: (Color online) Exponent m vs Ni concentration p for $Ni_pMg_{1-p}(OH)_2$. p_c $(= 0.1)$ and p_1 $(= 1/3)$ (denoted by arrows).

domains. If R is the characteristic domain size, the wall area grows as R^2 as the domains grow. Because the total number of spins is fixed, however, the number of domains decreases as R^{-3}. Thus the total number of spins in the walls decreases as R^{-1}. The magnetization from the domain walls in the FC state can be related to the domain size R,

$$M_{wall} \approx R^{-1}. \tag{4}$$

The field dependence of M_{wall} may be described by

$$M_{wall} \approx H^m, \tag{5}$$

at low temperatures, where $R \approx H^{-m}$ and m is the exponent for the domain size. Here we assume that the absorption χ'' may be given by

$$\chi'' \approx \frac{M_{wall}}{H} \approx H^{m-1} \approx H^{\zeta}, \tag{6}$$

with

$$m = \zeta + 1.$$

Since the exponent ζ can be determined experimentally, the exponent m can be obtained for each Ni concentration p.

Figure 16 shows the exponent m as a function of Ni concentration for $Ni_pMg_{1-p}(OH)_2$. The exponent m is determined as 3.12 ± 0.04 for $p = 1$, 2.96 ± 0.03 for $p = 0.80$, 2.47 ± 0.06 for $p = 0.50$, 2.28 ± 0.08 for $p = 0.315$, 2.20 ± 0.07 for $p = 0.25$, and 2.35 ± 0.07 for $p = 0.10$. It is found that m decreases with decreasing Ni concentration and takes a minimum around $p \approx p_1 = 0.33$, where p_1 is the estimated critical concentration when only the first

neighbor interaction is taken into account; $p_1 = 2/z_1 = 1/3$. Our results of m are on the same order as those for other dilute Ising random antiferromagnets; $m = 2.9 \pm 0.1$ for $Fe_{0.6}Zn_{0.4}F_2$ (Mattsson et al.[18]), $m = 3.2 \pm 0.3$ for $Fe_{0.46}Zn_{0.56}F_2$ (Ledermann et al.[17]), and $m = 2.7 \pm 0.1$ for $Fe_{0.5}Zn_{0.5}F_2$ (Feng et al.[19]). According to Mattsson et al.,[18] m can be expressed in terms of

$$m = \nu_H + (2D_s + 2 - 2d)/(d - 2), \tag{7}$$

where d is the spatial dimensionality of the system, ν_H is the exponent for the system with smooth domain walls, and D_s is the fractal dimension of the domain walls on short length scales. For $d = 3$, we have

$$m = \nu_H + (2D_s - 4). \tag{8}$$

The exponent ν_H can be evaluated from the neutron scattering experiment. If we assume that ν_H $(= 2.1)$[18] may be independent of the Ni concentration p, the fractal dimension D_s can be evaluated as $D_s = 2.5$ for $m = 3.12$ $(p = 1)$ and $D_s = 2.1$ for $m = 2.28$ $(p = 0.315)$. This means that the fractal dimension decreases from 2.5 to 2.1 with the dilution of Mg^{2+} ions. These value of D_s may be consistent with the theoretical predictions; $D_s = 2.27 \pm 0.02$ by Middleton and Fisher.[20]

B. Griffiths phase in the magnetic phase diagram

Figure 17 shows the magnetic phase diagram for $Ni_pMg_{1-p}(OH)_2$. The original magnetic phase diagram was reported by Suzuki et al.[14] The magnetic-field-induced Griffiths phase is plotted in this original magnetic phase diagram. Here the Griffith temperature T_G is defined by a peak temperature where the absorption χ'' exhibits a peak in the presence of H (typically $20 \leq H \leq 50$ kOe). It is found that T_G is strongly dependent on the Ni concentration p in our system. This result is rather different from our expectation before the measurements: T_G is independent of the concentration and coincides with the critical temperature of the undiluted system $(p = 1)$. For a ferromagnet with site or bond dilution, it is predicted that the Griffiths phase G is the region between the horizontal line $(T = T_G)$ and the phase boundary for the onset of ferromagnetism. The latter boundary meets the zero-temperature axis at the percolation threshold p_c. The Griffiths temperature T_G is the critical temperature of the undiluted system $(p = 1)$.[21]

For $p = 1$, 0.60, and 0.50, the Griffiths temperature T_G is higher than the corresponding Néel temperature. It decreases with increasing H for $H > 20$ kOe. This behavior may be due to the antiferromagnetic nature of the spin fluctuations. The effective interplanar antiferromagnetic interactions is expressed by $J'(\xi_a/a)^2$, where a is the in-plane lattice constant, ξ_a is the in-plane ferromagnetic spin correlation length, and J' is the antiferromagnetic interplanar interaction such as J_2 and J_3 in our system.

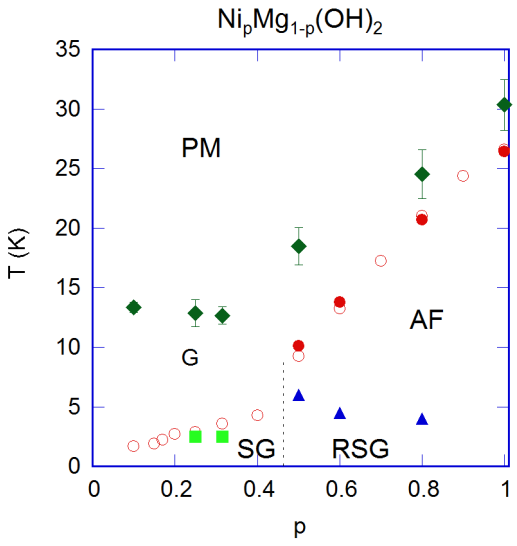

FIG. 17: (Color online) Magnetic phase diagram (critical temperature vs Ni concentration p) of $Ni_pMg_{1-p}(OH)_2$. PM: paramagnetic phase, AF: antiferromagnetic phase, SG: spin-glass phase, RSG: reentrant spin-glass phase, and G: Griffiths phase. The original magnetic phase diagram was reported by Suzuki et al.[14] (see the detail for the definition of open circle, closed square, closed triangle in the Reference[14]). The magnetic-field-induced Griffiths phase is plotted in this original magnetic phase diagram. Note that the Griffith temperature T_G (closed diamond) is a peak temperature where the absorption χ'' exhibits a peak in the presence of H (typically $20 \leq H \leq 50$ kOe). The bars of the data of T_G denotes the change of T_G as H is changed. In this sense, the Griffiths phase is the magnetic-induced phase. The data reported by Enoki and Tsujikawa[9] are denoted by open circles. The dotted vertical line $(p \simeq 0.46)$ is the phase boundary between the RSG phase and the SG phase.

This effective interplanar antiferromagnetic interaction becomes dominant near the Néel temperature through the dramatic growth of the in-plane ferromagnetic spin correlation length.

For $p = 0.315$, 0.25, and 0.10, T_G is higher than the spin glass freezing temperature T_{SG}. It is almost independent of p: $T_G \approx 13.7$ K. The Griffiths temperature slightly increases with increasing H. This behavior may be due to the ferromagnetic nature of the spin fluctuation. The effective interplanar antiferromagnetic interactions are much weaker than the intraplanar exchange interaction since there is only ferromagnetic short range order at any finite temperature below the percolation threshold $(p = p_1)$.

Finally we note that the existence of Griffiths phase has been confirmed by Deguchi et al.[16] in $Ni_pMg_{1-p}(OH)_2$ with $p = 0.42$ from the slow decay of thermoremnant magnetization M_{TRM}. The system is quenched from 50 K to a temperature in the presence of magnetic field H $(= 100$ Oe) (FC cooling) and annealed at a temperature T_0 for a wait time t_w $(= 10$ min). After the magnetic field H is reduced to zero at $t = 0$, M_{TRM} is measured

as a function of time. The slow relaxation of M_{TRM} is clearly observed in the Griffiths phase ($T_N < T_0 < T_G$); where $T_N \approx 7.5$ K and T_G (= 17 K). This value of T_G for $p = 0.42$ is a little lower than the value of T_G (= 14 K) evaluated from our magnetic phase diagram.

VI. CONCLUSION

The nature of the magnetic-field-induced Griffiths phase in 3D Ising random magnet $Ni_pMg_{1-p}(OH)_2$ (p = 0.10, 0.25, 0.315, 0.50, 0.80, and 1) has been studied from the absorption χ'' (the out-of phase in AC magnetic susceptibility) in the field-cooled (FC) state. The Griffiths temperature T_G is defined as the peak temperature of χ'' vs T above $T_N(H = 0)$. The peak height $\chi''(T_G)$ drastically increases with increasing H for $H > 20$ kOe, following a power form, $\chi''(T_G) \approx H^{m-1}$. The exponent m depends on the Ni concentration; $m = 3.12 \pm 0.04$ for $p = 1$ and 2.28 ± 0.08 for $p = 0.315$.

Acknowledgments

We are grateful to Prof. Toshiaki Enoki for providing us with samples of $Ni_pMg_{1-p}(OH)_2$ and for his invaluable suggestions and discussions.

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