Griffiths phase and its evolution with Mn-site disorder in the half-doped manganite $\Pr_{0.5}$ $\Pr_{0.5}$ Mn_{1-v} Ga_vO_3 ($y=0.0$, 0.025, and 0.05)

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We show Griffiths phase (GP)-like behavior in half-doped $Pr_{0.5}Sr_{0.5}MnO_3$ having ferromagnetic (FM) spin correlation above FM transition (T_C) without any spontaneous magnetization. Temperature range and exponent of GP increase with Ga substitution at Mn site which is not related with structural disorder as Ga does not affect the structure, moreover, reduces Jahn-Teller active Mn^{3+} ions. FM clusters both above and below T_c reduces their size with Ga. Hence, it is considered that GP originates from phase inhomogeneity and evolves with addition of disorder due to change in its length scale.

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I. INTRODUCTION

The phase inhomogeneity and the effect of quenched disorder in manganite with generic formula $R_{1-x}A_xMnO_3$ where *R* is trivalent rare earth and *A* is divalent alkalineearth ion) have attracted considerable interests.¹ Among the various forms of inhomogeneous phases, preformation of ferromagnetic (FM) clusters much above the FM long-range ordering temperature (T_C) is a well accepted phenomenon.^{1[,2](#page-4-1)} This often leads to Griffiths singularity³ originally proposed for randomly diluted Ising FM where only a fraction of the lattice sites are occupied with spins and the rest are either vacant or filled with nonmagnetic atoms (without spin). Thus the nearest-neighbor exchange bonds with strength *J* and 0 are distributed randomly with probability *p* and $(1-p)$, respectively. For $p < p_c$ (percolation threshold) no long-range FM order is established. However, for $p \ge p_c$, long-range FM interaction sets in at $T_c(p)$ which is below the ordering temperature of undiluted or homogeneous system $T_c(p=1)$ and the latter is known as Griffiths temperature (T_G) . In the temperature region $T_c(p) < T < T_c$, the system is considered to exist in the Griffiths phase (GP) which neither exhibits pure paramagnetic (PM) behavior nor long-range FM order. In this temperature range, though the system does not hold spontaneous magnetization, there exists spatially distributed regions that are devoid of disorder forming finitesize clusters having ferromagnetically correlated spins and the magnetization fails to be analytic. Bray⁴ generalized this argument for any bond distribution (instead of bonds having strength of only J and 0) which reduces the long-range ordering transition temperature. This generalization becomes helpful to reckon GP-like properties in variety of systems including manganite, $5-10$ $5-10$ cobaltite, 11 and many other compounds[.12](#page-4-7)[–15](#page-4-8)

Quenched disorder is prerequisite for formation of GP. Manganites are intrinsically disordered because of the random distribution of cations of different sizes and valences at the R/A site. Along with the local lattice distortion contributed by Jahn-Teller (J-T) active Mn^{3+} ions, this size mismatch of *R*/*A* ions also induces variation in the Mn-O-Mn bond angle and length, translating it to the case of bond disorder for the magnetically active Mn-O-Mn network. Moreover, additional disorder can be introduced through chemical substitution at two different crystallographic sites, viz., *R*/*A* site or Mn site. Both these have distinct effects. The variation in R/A atoms causes (i) modification of the structure or the tolerance factor (τ) , (ii) change in the variance (σ^2) of ionic radii, and (iii) modulation of Mn^{3+}/Mn^{4+} ratio.¹⁶ The bond distribution is affected by all these factors. Most of the recent studies related to GP in manganites are on the *R*/*A*-site disorder. Though they emphasize on structural disorder, contributed by different factors, there appears to be lack of consensus on the exact source of disorder responsible for the observation of $GP^{5,6,9,10}$ $GP^{5,6,9,10}$ $GP^{5,6,9,10}$ $GP^{5,6,9,10}$ $GP^{5,6,9,10}$ For examples, it is considered that the disorder arising from the bending of Mn-O-Mn bond induces GP in $La_{1-x}Ca_xMnO_3(x=0.3).^{5,9}$ $La_{1-x}Ca_xMnO_3(x=0.3).^{5,9}$ $La_{1-x}Ca_xMnO_3(x=0.3).^{5,9}$ $La_{1-x}Ca_xMnO_3(x=0.3).^{5,9}$ On the contrary, static J-T distortion acts as quenched disorder for formation of GP in $La_{1-x}Sr_xMnO_3$.^{[6](#page-4-10)} Yet, disorder arising from the size variance of R/A -site atoms is found to be responsible for the development of GP in $La_{1-x}Ba_xMnO_3$ series.¹⁰ In this respect, Mn-site substitution of nonmagnetic ions having similar radii of Mn^{3+} or Mn^{4+} offers a particular way to introduce disorder in the system. The significant consequences of this substitution is that there is no structural distortion induced through the variation in τ and/or σ^2 . Thus such substitutions can lead to random dilution (site) of the magnetic lattice, making the associated bonds of strength *J* = 0 and almost mimic the original model of Griffiths.

Here we study the GP-like phase in Pr_0 , Sr_0 , MnO_3 (PSMO) and its progressive development on addition of quenched disorder at the Mn site by substitution of nonmagnetic $Ga^{3+}(d^{10})$. On cooling, the parent compound PSMO undergoes second-order PM to FM transition followed by first-order FM to antiferromagnetic (AF) transition.¹⁷ The FM T_c decreases linearly with Ga substitution, however, there is no significant change in structural parameters because of the matching ionic radii of Mn^{3+} and Ga^{3+} .^{[18](#page-4-13)} Though the PM to FM transition in PSMO is characterized to be second order,¹⁹ the phase immediately above T_c is not pure PM.¹⁸ Here it is shown that this inhomogeneous phase above T_c has characteristics similar to GP. Presence of FM clusters in this temperature range is inferred from the second-order ac susceptibility (χ_2) but they do not have spontaneous magnetization (M_S) . It is rather significant that both the temperature range and the strength of the GP increases progressively with the addition of quenched disorder

FIG. 1. (Color online) Temperature variation in inverse dc susceptibility (χ_{dc}^{-1}) measured in 0.1, 0.5, 1, and 10 kOe field are plotted for parent compound $(y=0)$. In inset inverse ac susceptibility (χ_{ac}^{-1}) measured in ac field 2 Oe and frequency 9 Hz is plotted as a function of temperature for the same sample.

in the magnetic lattice by Ga substitutions without any change in *R*/*A*-site constituents or in τ and/or σ^2 .

II. EXPERIMENTAL DETAILS

Samples of Pr_{0.5}Sr_{0.5}Mn_{1-*y*}Ga_{*y*}O₃ series with *y*=0.0, 0.025, and 0.05 are studied here. They are prepared by standard solid-state ceramic route and characterized by various techniques such as Rietveld profile refinement of x-ray diffraction data, Iodometric titration, microprobe analysis, etc[.18](#page-4-13) dc-magnetization (DCM) data are collected with vibrating sample magnetometer (PPMS-VSM, Quantum Design). ac susceptibility is measured with homemade ac-susceptibility (ACS) setup.²⁰ Some of the magnetic data are also crosschecked in a superconducting quantum interference device (SQUID) magnetometer (MPMS, Quantum Design). The T_C for all the samples are deduced from the inflection point of low-field ac-susceptibility data. In case of the parent compound, this is slightly higher than the T_c found from the analysis of critical isotherms.¹⁹

III. RESULTS AND DISCUSSIONS

The downturn in inverse susceptibility (χ^{-1}) as a function of temperature above T_C is considered to be a hallmark of Griffiths singularity.^{5[,6,](#page-4-10)[13](#page-4-16)} The temperature dependence of χ^{-1} deduced from dc magnetization (M) measured in different fields for the parent compound $(y=0)$ of the series Pr_{0.5}Sr_{0.5}Mn_{1−y}Ga_yO₃ (PSMO) is shown in Fig. [1.](#page-1-0) The inset of Fig. [1](#page-1-0) shows the χ^{-1} vs *T* found from the ac susceptibility measured in ac field 2 Oe and frequency 9 Hz. In high T , χ^{-1} varies linearly with T indicating Curie-Weiss (CW) behavior. However, with the decrease in *T*, sharp downturn is observed much above T_C in $\chi^{-1}(T)$ for the measurements done in both ac field and lower dc fields indicating nonanalytic behavior of magnetization arising from Griffiths singularity. This downturn in $\chi^{-1}(T)$ is an important observation which distinguishes the Griffiths singularity from smeared phase transition because the latter case gives rise to an upward curvature in $\chi^{-1}(T)$ above T_C , deviating from CW behavior.^{2[,21](#page-4-17)} The softening of the downturn in χ^{-1} with the progressive in-

FIG. 2. (Color online) (a) Second-order ac susceptibility (χ_2) for *y*= 0 sample is plotted as a function of temperature. Inset shows the magnified view of data above T_C . (b) Arrott plot $(M^2 \text{ vs } H/M)$ of isotherms collected at different temperatures both below and above T_c are plotted. Lines are due to straight-line fitting of plot in high field.

crease in field from 0.1 0.1 to 10 kOe (Fig. 1) is a typical signature of the GP which has also been observed in variety of other systems[.6](#page-4-10)[,13,](#page-4-16)[14](#page-4-18) It is clear from Fig. [1](#page-1-0) that in 10 kOe field, χ^{-1} becomes almost straight in GP region resembling CW behavior. This indicates that because of the linear increase in *M* with the field for the PM, in high fields the contribution of FM clusters to the total magnetic susceptibility of the system is dominated by that of PM matrix.

The basic characteristics of GP regime is that above T_C there exist finite-size clusters with FM correlated spins. However, the system as a whole would not develop static long-range order and M_S is zero which is opposite in case of smeared phase transition.²² We have confirmed these two aspects from the studies of $\chi_2(T)$ and Arrott plot. As such there is no higher order ac susceptibilities in the PM state and they appear when the spin correlation develops in the system. In the absence of external dc field, χ_2 arises due to symmetry-breaking field originating from the ferromagnetically correlated spins of clusters. Hence, the appearance of χ_2 unambiguously indicate the presence of spin clusters with FM correlation. Figure $2(a)$ $2(a)$ shows χ_2 vs *T* measured in the ac field of 9 Oe and frequency 131 Hz, without any external dc field for PSMO sample. The sharp negative peak in χ_2 is associated with T_C of the sample. However, χ_2 appears much above T_C [inset of Fig. [2](#page-1-1)(a)] indicating the formation of FM correlated spin clusters. It may be noted that with the decrease in *T*, magnitude of χ_2 increases slowly and shows a sharp change at a characteristic temperature, T_C^R (discussed later), before the peak around T_C . To check if the observed χ_2 is associated with the M_S , we have collected magnetic isotherm data $(M-H)$ at different *T* both below and above T_C . In Fig. [2](#page-1-1)(b) we show the plot of M^2 vs H/M (only a few such isotherms are shown for clarity), which is known as Arrott plot.^{[23](#page-4-20)} Intercept on positive M^2 axis of the high-field extrapolation of this plot gives M_S . It is clear from Fig. [2](#page-1-1)(b) that there is no M_S in the temperature region (above T_C) where finite χ_2 χ_2 is observed. Thus Figs. [1](#page-1-0) and 2 unambiguously show that in PSMO, the phase above T_c is GP and not pure PM.

Now we study the evolution of GP as additional quenched disorder is introduced in the parent compound of the Pr_{0.5}Sr_{0.5}Mn_{1→}_{*y*}Ga_{*y*}O₃ series, through the Ga substitution at the Mn site. Figure $3(a)$ $3(a)$ shows the temperature dependence

FIG. 3. (Color online) (a) Temperature variation in inverse dc susceptibility (χ_{dc}^{-1}) measured in 0.1 kOe is plotted for *y*=0.025 and 0.05 samples of $Pr_{0.5}Sr_{0.5}Mn_{1-y}Ga_yO_3$ series. (b) Susceptibility (χ) data for all the samples are plotted as a function of temperature following Eq. (1) (1) (1) in double-logarithmic scale. Continuous lines are due to straight-line fittings. Plot for *y*= 0.025 sample is shifted downward by −0.35 for clarity.

of χ^{-1} deduced from dc *M* measured in 0.1 kOe field for *y* $= 0.025$ and 0.05 samples of the series showing similar downturn as in Fig. [1.](#page-1-0) Moreover, the magnetic phase of both these samples above their respective $T_{\rm C}$'s have qualitatively similar features observed in PSMO i.e., presence of χ_2 without any M_S . Usually, Griffiths singularity is characterized by the χ^{-1} exponent (λ) lower than unity, as obtained from following relation:²⁴

$$
\chi^{-1} = (T - T_C^R)^{1 - \lambda},\tag{1}
$$

where $0 \leq \lambda < 1$ and T_C^R is the critical temperature of random FM where susceptibility tend to diverge. It is pertinent to note here that in an earlier study it was proposed that disorder can give rise to divergence in susceptibility.²⁵ It is shown from the theoretical studies on simple model systems that in Griffiths phase this divergence gives rise to the power-law behavior of the form of Eq. $(1).^{24,26}$ $(1).^{24,26}$ $(1).^{24,26}$ $(1).^{24,26}$ $(1).^{24,26}$ $(1).^{24,26}$ However, in the recent time, this power-law behavior given in Eq. (1) (1) (1) is extensively used to characterize the Griffiths singularity in the experimental studies on variety of complicated systems[.5,](#page-4-4)[9,](#page-4-11)[10](#page-4-5)[,13](#page-4-16)[–15](#page-4-8) This power-law relation appears to be a modified form of CW law where the exponent λ signifies deviation from the CW behavior. In the pure paramagnetic region λ is expected to be zero. The Griffiths phase is characterized by the presence of "finite-size" FM clusters within PM matrix above T_C where susceptibility deviates from CW behavior and the fit-ting of Eq. ([1](#page-2-1)) yields finite value of λ_{GP} (between 0 and 1). Thus it is clear that higher the value of λ_{GP} stronger is the deviation from CW behavior.

To find out λ , χ^{-1} vs $T/T_C^R - 1$ is plotted on \log_{10} - \log_{10} scale and the slope of fitted straight lines $[Eq. (1)]$ $[Eq. (1)]$ $[Eq. (1)]$ both in GP regime and PM state gives λ_{GP} and λ_{PM} , respectively. It can be mentioned that incorrect value of T_C^R in Eq. ([1](#page-2-1)) can lead to unphysical fitting and erroneous determination of λ . To estimate T_C^R accurately we have followed a method where initially λ_{PM} is calculated with $T_C^R = T_C$. We then adjusted the T_C^R in above fitting and accepted the value for which λ_{PM} becomes close to zero. This is done following the fact that GP transforms into conventional PM state above T_G and system obeys the CW law. With this T_C^R , we have calculated λ_{GP} . The T_G is calculated from the onset of downturn in $\chi^{-1}(T)$.^{[13](#page-4-16)}

TABLE I. Characteristic temperatures, the inverse susceptibility exponents (λ) , and the range of Griffiths phase defined as GP =*TG*−*TC*-/*TC*100 for the series Pr0.5Sr0.5Mn1−*y*Ga*y*O3. DCM and ACS represent dc magnetization and ac susceptibility, respectively.

Ga(y)	0%	2.5%	5.0%
$T_C(K)$	269.2	236.4	201.2
$T_G(K)$	315	300	292
$T_C^R(K)$	276.2	250.1	226.1
$\lambda_{PM}(DCM)$	0.002(4)	0.008(3)	0.006(1)
$\lambda_{GP}(DCM)$	0.72(3)	0.74(1)	0.80(2)
$\lambda_{GP}(ACS)$	0.69(4)	0.75(3)	0.85(1)
$GP(\%T_C)$	17	26.9	45.1

Such fitting of Eq. (1) (1) (1) is shown in Fig. $3(b)$ $3(b)$ for all the three samples. The λ_{GP} is calculated from both DCM data as well as ACS data for all the samples and the values are given in Table [I](#page-2-2) along with T_C , T_C^R , and T_G values. It is observed that λ_{GP} estimated from DCM and ACS data are reasonably close. Moreover, $\lambda_{GP}(\text{DCM})$ is found to be consistent and reproducible as calculated from the magnetization data collected in different instruments (VSM and SQUID). It needs to be mentioned that Eq. (1) (1) (1) is proposed from the theoretical studies on the Griffiths-type singularity on rather simple models.^{24[,26](#page-4-23)} Even in the original work of Griffiths, it is envisaged that the susceptibility would tend to diverge before the onset of long-range order at T_c .^{[3](#page-4-2)} This is clearly indicated in an earlier work, 27 contemporaneous to the original work of Griffiths. In this pioneering work of McCoy, a clear word of caution is given for the characterization of Griffiths-type singularity from the measurement of susceptibility in nonzero field because of possible appearance of spontaneous magnetization at temperatures above the long-range FM ordering at T_c . This indicates that the log_{10} -log₁₀ plot of the inverse of susceptibility, measured in finite field, as a function of temperature would tend to deviate from straight line in the GP regime as one approaches the T_C from high-temperature side. This would require a correction to the power-law relation in the form of Eq. (1) (1) (1) for realistic systems, which are rather complicated. However, it is somewhat intriguing that in Fig. $3(b)$ $3(b)$, the log₁₀-log₁₀ plot of susceptibility data measured in finite field fall on straight lines in the GP region (immediately above T_C) even for this series of samples, which is very complicated because of the intrinsic electronic phase separation both above and below T_c . This signifies that, in the first approximation, Eq. (1) (1) (1) has wider validity.

The obtained value of λ_{GP} for parent compound is com-parable with other manganites.^{5,[9,](#page-4-11)[10](#page-4-5)} It is clear from above discussions that large value of λ_{GP} indicates that Griffiths singularity is reasonably strong in PSMO. Moreover, with the substitution of Ga, λ_{GP} increases indicating further increase in the GP properties. It is rather significant that though the T_c decreases linearly, the temperature range over which the GP exist increases almost linearly with Ga substitution. It is clear from Table [I](#page-2-2) that the temperature range of GP normalized with the respective T_c 's calculated as $GP(\mathcal{C}T_c)$ $=[(T_G-T_C)/T_C] \times 100$ increases almost linearly with the addition of quenched disorder.

The presence of GP in half-doped PSMO is quite significant as in La-based manganites GP is usually observed in relatively low hole doped $(x \le 0.3)$ compositions.^{6,[9](#page-4-11)[,10](#page-4-5)} As such, theoretically it is proposed that around half doping in manganites the disorder arising from local lattice distortion is less likely to give rise to a inhomogeneous state and GP[.28](#page-4-25) The PM and FM phases in PSMO being of same structure,²⁹ crystallographic strain related disorder may not be important here. However, experimentally, PSMO is found to be inhomogeneous both above and below the FM transition.¹⁸ Additionally, considerable size mismatch of R/A ions (Pr^{3+}) $= 1.29$ Å and $Sr^{2+} = 1.44$ Å) as well as departure from the undistorted cubic structure would quench the bond disorder in this intermediate bandwidth system. This system has other intriguing features which may have serious consequences on the formation of GP. The ground state of this compound is at the phase boundary of AF and FM phases but it has AF state at low *T* and an unconventional FM state at higher *T*. Though the PM to FM phase transition is second order, it does not belong to any known universality class and the magnetic interaction appears to be rather complicated both in terms of range and dimensionality.¹⁹

Notwithstanding this complicated scenario, addition of quenched disorder in the form of Ga at the Mn site of PSMO has some interesting consequences vis-à-vis the La-based systems with R/A site disorder. Though T_G decreases slowly, both the temperature range and strength of the GP is en-hanced with Ga (Table [I](#page-2-2)). It may be noted that small amount of Ga substitution in this series introduces quenched disorder by simply diluting the magnetic lattice giving rise to a linear decrease in T_c indicating a mean-field-like situation.¹⁸ This substitution does not add any structural distortion to the original system. On the contrary, it is rather significant that Ga substitution reduces the concentration of J-T active Mn^{3+} ions[.18](#page-4-13) Hence, it may be ruled out that the additional structural disorder is the cause of the enhancement of GP. Thus, random substitution of Ga emulates a situation where disorder is introduced in the form of simple dilution of the magnetic lattice rendering the related bond strength (J) to zero. This would imply that the evolution of GP phase on progressive dilution of magnetic lattice should have different behavior compared to the recent studies where disorder was introduced in manganites through R/A -site substitution.^{6[,10](#page-4-5)} Manifestation of this difference probably becomes evident with the decrease in T_G in this system. Though such decrease in T_G is not predicted in the simplified model of Griffiths,³ we try to justify that it may be intrinsic to such a electronically phase separated systems and needs further investigation. In the classic work of Imry and $Ma₃₀$ it was argued that random quenched disorder can destabilize the long-rangeorder system favoring formation of finite-size clusters. Recently, it is shown that the FM cluster size decreases with the increase in the random disorder in the form of Ga in PSMO.¹⁸ Thus we propose that the decrease in T_G with disorder (Ga) arises from the decrease in the cluster size because spin correlation in smaller clusters will be more susceptible to thermal fluctuations shifting the onset of GP to lower $T²²$ $T²²$ $T²²$

It has been stated earlier that the exponent λ_{GP} associated with inverse susceptibility is a measure of the deviation from CW law indicating that it is related to the short-range spin clusters in the temperature range between T_c and T_c . In the theoretical studies λ_{GP} has a correlation with the size distri-bution of the spin clusters in the system.^{5,[24–](#page-4-21)[26](#page-4-23)} Here, we argue that in the present system, as the size of the FM infinite clusters deceases in FM phase below T_c with the increase in disorder¹⁸ the length scale of the finite-size spin clusters also decreases above T_c . This would naturally increase the number of finite-size spin clusters which is a probable cause for the enhancement of the GP properties. Moreover, with the increase in disorder or decrease in the length scale of spin clusters, T_c at which the infinite long-range order is established will be shifted further below the T_G exhibiting GP over a wider T range.⁵ Therefore, it appears that as the system becomes more inhomogeneous in shorter length scale with the Ga substitution, the GP properties enhance in PSMO with respect to both its temperature range and associated exponent (λ_{GP}) . Significantly, it is shown from a Monte Carlo study that the strength of the GP increases monotonically with the decrease in the dimensionality of the system. 25 This may have some correspondence with the present system and prompt further investigation on these intrinsically phase separated manganites.

IV. CONCLUSION

In conclusion, we show reasonably pronounced Griffiths phase-like behavior in half-doped Pr_0 , Sr_0 , MnO_3 . In this GP regime there exist FM correlation of spins without spontaneous magnetization. Both, temperature range and associated exponent (λ_{GP}) characterizing the GP increases with the addition of disorder in the magnetic lattice in the form of substitution of Ga at the Mn site. This enhancement of GP properties is not directly related to structural effects since Ga predominately replaces J-T active Mn^{3+} ions and does not have any significant effect on lattice parameters. However, Ga directly introduces disorder in the form dilution of the magnetic lattice and makes the associated bond strength *J* = 0. The parent compound, PSMO, is shown to be inhomogeneous having FM clusters both above and below its second-order PM to FM transition. Addition of disorder in the magnetic lattice further reduces the size of these magnetic clusters, which helps in enhancement of GP properties. Thus it appears that the magnetic inhomogeneity and decrease in its length scale with additional disorder (Ga) is the primary cause of the appearance of GP regime and its subsequent enhancement in this series. We believe that these results have broader significance and would provoke both theoretical as well as experimental studies especially on other low doped or optimally doped manganites with similar Mn-site disorders.

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