PHYSICAL REVIEW B **76**, 092404 (2007)

Extreme sensitivity of the Griffiths phase to magnetic field in single crystal $La_{0.73}Ba_{0.27}MnO_3$

Wanjun Jiang,* X. Z. Zhou, and Gwyn Williams
Department of Physics and Astronomy, University of Manitoba, Winnipeg, Manitoba, Canada R3T 2N2

Y. Mukovskii and K. Glazyrin
Moscow State Steel and Alloys Institute, Moscow 119049, Russia

(Received 8 June 2007; published 12 September 2007)

Field-dependent ac susceptibility and low field (<150 Oe) magnetization measurements on single crystal $La_{0.73}Ba_{0.27}MnO_3$ reveal features consistent with the appearance of a Griffiths phase in the temperature interval between $T_C=245$ and 345 K. Such features, however, are suppressed by fields of only 150 Oe, a result consistent with reduced value of the susceptibility "exponent" (y) deduced from these data. Comparisons with the behavior of other doped manganites near optimal doping suggest that the proximity of the mean A-site radius $\langle r_A \rangle$ to its undistorted value r_A^0 *might* provide an appropriate measure of the tendency of these systems to nucleate a Griffiths phase.

DOI: 10.1103/PhysRevB.76.092404 PACS number(s): 75.40.Cx, 75.47.Lx

There is a growing body of evidence supporting the occurrence of a Griffiths phase not only in doped Mn perovskites[1,2]—which appear particularly susceptible to the influence of disorder[3]—but also in a variety of other systems containing rare-earth[4] and transition metal ions.[5]

The original problem considered by Griffiths[6] was percolative in nature, one in which nearest neighbor (exchange) bonds characterized by strength J occur with probability p; otherwise, the bond strength is zero [with probability $(1-p)$]. For $p<p_C$ (the percolation probability), there is zero probability of forming an infinite percolating "backbone"—equivalently, the correlation length does not diverge—and thus, as expected, no cooperative ferromagnetic transition occurs. However, for $p>p_C$, ferromagnetism is established, albeit at a temperature $T_C(p)$ somewhat below that of the undiluted system $[T_C(p=1)=T_G]$. The Griffiths phase appears in the temperature interval $T_C(p) \leqslant T \leqslant T_G$. In this regime, the behavior of the system is neither that of a simple paramagnet nor is an infinite percolating backbone established; rather, the response is determined by a series of clusters and/or correlated volumes of finite extent. Specifically, this response is dominated by those correlated regions and/or clusters with the largest susceptibilities. In close proximity to $T_C(p)$, the system can be "unblocked" by application of an applied field which causes the cluster size—in current critical phenomenon language, the correlation length—to diverge. Correspondingly, there is a sharp increase in the magnetization, a feature considered as a defining characteristic of a Griffiths singularity, a singularity in the sense that the free energy—and consequently its derivatives (the experimentally measured response functions)—is nonanalytic over the associated temperature interval. While this behavior of the magnetization is reminiscent of a first order/discontinuous transition, the phase change at $T_C(p)$ is continuous, with the corresponding power-law dependence of the magnetization on field along the critical isotherm:

$$M \propto H^{1/\delta}, \qquad (1)$$

yielding large (~20) values[1] for the "exponent" δ. A number of generalizations have been made to Griffiths' original approach[7] within which the overall behavior outlined above persists.

To return to the manganites, the Griffiths phase approach was shown to provide not only a consistent description of available data on $La_{0.7}Ca_{0.3}MnO_3$ but also a framework for discussing the variation of magnetic and transport properties with dopant species Pb, Sr, and Ca near optimal doping.[1] In general terms, as the average ionic radius of these A (La) site substituted ions falls, so does the disorder-corrected[8] transition temperature $T_C(\langle r_A \rangle, 0)$; more importantly, the variance σ in the A-site ionic radii, a measure of this disorder, plays a pivotal role.[1,8] An exception noted[1] to this general behavior occurred in single crystal $La_{0.72}Ba_{0.27}MnO_3$—again near optimal doping—which was reported as having $T_C=245$ K, intermediate between its Ca- and Sr-doped single crystal counterparts, but more importantly with a T_C below that of a Ca-Pb-doped sample which displayed evidence of Griffiths behavior. This Ba-doped single crystal displayed a conventional continuous paramagnetic to ferromagnetic phase transition[9] governed not just by Heisenberg model exponents[10] but also with critical amplitudes consistent with the same model. These conclusions were confirmed by more recent measurements of the field-dependent ac susceptibility.[11] In particular, no indication of the occurrence of a Griffiths phase was evident in either series of measurements. This Brief Report presents *low* field ac susceptibility and magnetization data on this same Ba single crystal, which, while revealing the appearance of a Griffiths phase in it, indicate subtle differences with data acquired from its Ca-, Pb-, and Sr-doped counterparts.

Simultaneous measurements of the ac susceptibility (at 1 kHz with a driving field amplitude of 100 mOe rms, in static fields between zero field and 150 Oe) and the corresponding magnetization were carried out in a Quantum Design PPMS model 6000 magnetometer and/or susceptometer. Within experimental uncertainty (typically ±0.5 K), these data display no thermal hysteresis above the ferromagnetic ordering temperature. All fields were applied along the largest dimension of this 0.163 g semicylindrical rod of length 6 mm and average radius 1.6 mm; the preparation and various characteristics of this single crystal have been discussed previously.[9] In particular, this single crystal displayed a mo-

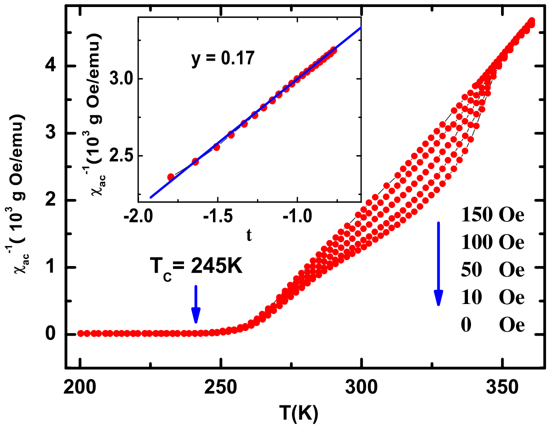

FIG. 1. (Color online) The inverse ac susceptibility, measured on warming following zero-field cooling under various (nominal) applied fields. The inset reproduces the zero-field data on a log-log scale, with $t=(T-T_C^*)/T_C^*$ and $T_C^*=261$ K; this form tests the power-law, Eq. (2), and yields $y \sim 0.17$.

saicity of less than 1°, and while no direct means of assessing the oxygen content is available to us, the measured low temperature saturation magnetization[9] indicates that this specimen is close to being stoichiometric. Relevant data have been corrected for background and demagnetization effects, with a demagnetization factor estimated from both[9] the maximum zero-field susceptibility and from shearing curves close to T_C. At each measuring field and temperature, appropriate waiting times were adopted to ensure that equilibrium conditions prevailed. The relevant data are presented in Figs. 1 and 2.

In assessing the present data and comparing them with previous reports,[9,11] it is important to recall that conventional critical analyses of data on this[9] and related systems[12] are usually based on the use of an Arrott-Noakes[13] equation of state and involve extrapolations from the "technically saturated" regime. The latter is necessary for two principal reasons: first, to eliminate *noncritical* contributions to the measured response (arising from sources such as domain wall motion and coherent domain rotation), and second, since samples are rarely perfectly ellipsoidal in shape, to exclude data for which the corresponding demagnetizing field correction represents a significant fraction of the applied field (i.e., with $H_i=H_a$-NM, in the usual notation, this approach suppresses data with large uncertainties in the internal field H_i). This eliminates data obtained typically[9,12] in applied fields $(H_a) \leqslant 500$ Oe. Similarly, while critical analyses of ac susceptibility data can be carried out in fields as low as 0.1 Oe in the most favorable cases,[14] the manganites[11,15] (and indeed, pyrochlores[16]) are well known to exhibit a technical hardness far in excess of that which might be inferred from measured coercive fields; this limits critical analysis of ac susceptibility data on the present single crystal to fields exceeding[11] 20 kOe. These points are relevant, since manifestations of a Griffiths phase are known to be suppressed as the field is increased.[1,4] Such a result is expected[11] as the applied field is the appropriate conjugate field for uniform ferromagnetism; increasing the magnitude of the applied field thus promotes the growth of uniform ferromagnetism at the expense of its nonuniform Griffiths phase counterpart.

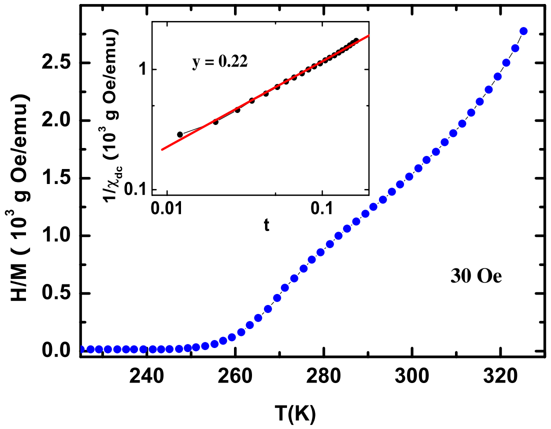

FIG. 2. (Color online) The inverse dc susceptibility measured on warming in a nominal field of 30 Oe after zero-field cooling. The inset replicates that in Fig. 1 and yields $y \sim 0.22$.

The data shown in Fig. 1 are significant in two aspects.

(i) They indicate an extreme sensitivity to applied (static) fields. The signature Griffiths phase response—a depression of χ^{-1} vs T curves below the Curie-Weiss line[1,4]—is completely quenched in fields of only 150 Oe in this system (such fields, incidentally, are well below the minimum values used in previous critical analyses of either magnetization or ac susceptibility data). An anticipated corollary is that the Griffiths singularity is correspondingly weak in this Ba-doped single crystal, with the inset in Fig. 1 yielding an estimate for the susceptibility exponent y deduced from the power-law form attributed to the inverse susceptibility in the Griffiths regime, viz.[1,2,4]

$$\chi^{-1} \propto (T-T_C^*)^{1-y} \quad (y>0) \tag{2}$$

of only $y \sim 0.17$ with $T_C^*=261$ K. Values reported for doped manganites are typically[1,2] $y=0.6-0.7$ but can sometimes be smaller in other systems,[4,5] 0.3–0.8.

(ii) These data revert to near Curie-Weiss-like behavior not only *above* $T_G \approx 345$ K but also in the region *immediately* above T_C (hence, the low value for y), unlike the response observed to date in other systems.

The corresponding low-field inverse magnetization data, Fig. 2, are qualitatively consistent with the ac susceptibility data, showing deviations away from Curie-Weiss behavior with a curvature comparable to that shown in Fig. 1. However, as ac susceptibility measurements can be carried out reliably to some 30 K higher than magnetization studies in the present measuring system and are not as sensitive to trapped field uncertainties, the estimate of the parameter y from the former data is likely more reliable. Nevertheless, these low field data reveal the occurrence of a Griffiths phase in a near optimally doped Ba single crystal. Here, however, this phase displays an extreme sensitivity to applied field, at least in regard to the measured T_C's of a number of doped Mn perovskites. Specifically, the data on the doped Mn perovskites reported in Ref. 1 showed that with decreasing T_C, these systems displayed enhanced Griffiths phase character-

TABLE I. Parameters characterizing some optimally doped Mn perovskites, $La_{1-x}A_xMnO_3$.

A	x	$\langle r_A \rangle$ (Å)	σ (Å)	T_C (K)	T_C $(\langle r_A \rangle, 0)$ (K)
Ca	0.3	1.205	0.0165	218	268
Ba	0.27	1.285	0.1128	245	523
Ca-Pb	0.34	1.242(5)	0.2778	286	434
Sr	0.3	1.244	0.0431	360	440

istics; additionally, these authors stressed the role played by both the average A-site radius $\langle r_A \rangle$ and its variance σ in determining the behavior of these systems, a point reiterated by a number of other studies.[8,17] The bending of the Mn-O-Mn bond accompanying decreases in $\langle r_A \rangle$ suppresses double exchange, with a consequent[18] decrease in T_C. However, the depression of T_C at constant $\langle r_A \rangle$ as σ increases suggests that local—rather than average—fluctuations predominate. In this context, the behavior of the Ba single crystal is anomalous. This can be seen from the comparisons summarized in Table I, which reproduces the values of $\langle r_A \rangle$, σ, and the measured ordering temperature T_C for the present sample and those reported in Ref. 1 The correlation between T_C and the Griffiths phase characteristics mentioned above is clearly not universal. To examine this correlation in greater detail, Table I also lists the disorder-corrected ordering temperatures $T_C(\langle r_A \rangle, 0)$ for the samples in question. Rodriguez-Martinez and Attfield[8] have proposed a semiquantitative model which provides a considerably unified description of the behavior of doped Mn perovskites (and indeed cuprate superconductors[19]) in the form of a comprehensive $T_C(\langle r_A \rangle, 0)$ vs $\langle r_A \rangle$ phase diagram. This was achieved through the use of a disorder-corrected ordering temperature $T_C(\langle r_A \rangle, 0)$ estimated from

$$T_C(\langle r_A \rangle, 0) = T_C(r_A^0, 0) - \alpha(r_A^0 - \langle r_A \rangle)^2. \tag{3}$$

In this expression, r_A^0 represents the ideal radius of the undistorted cubic perovskite structure (taken as 1.30 Å near optimal doping), so that the last term in Eq. (3) measures the departure of the actual structure from "ideal," i.e., a measure of the strain field. $T_C(r_A^0, 0)$ and α were proposed as universal constants for these optimally doped perovskites, assuming values of 350 K and 29×10^3 K Å^{-2}, respectively, from fitting available data. As all samples considered in Table I are close to optimal doping, this choice for the ideal radius r_A^0 is appropriate. As can be seen from this table, there is no clear correlation between the occurrence of Griffiths phase behavior and these tabulated parameters (or indeed the combination $\langle r_A \rangle / \sigma$) *except* that $\langle r_A \rangle$ for the present Ba sample is closest to r_A^0 of the *undistorted* structure mentioned above. Hence—from Eq. (3)—$T_C(\langle r_A \rangle, 0)$ is highest for this Ba-doped single crystal, exceeding that of its Sr-doped counterpart which displays little evidence of Griffiths behavior. Whether the latter, i.e., $r_A^0 - \langle r_A \rangle$ (and the accompanying strain field[8]), rather than the variance σ is thus a more appropriate measure of the "disorder"—and hence the tendency to nucleate a Griffiths phase—will need further study. The present data, nevertheless, confirm the continued subtleties displayed by CMR manganites.

Support for this work by the Natural Sciences and Engineering Research Council of Canada (NSERC) is gratefully acknowledged.

---

*jiang@physics.umanitoba.ca

[1] M. B. Salamon, P. Lin, and S. H. Chun, Phys. Rev. Lett. **88**, 197203 (2002); M. B. Salamon and S. H. Chun, Phys. Rev. B **68**, 014411 (2003); P. K. Chan, N. Goldenfeld, and M. B. Salamon, Phys. Rev. Lett. **97**, 137201 (2006).

[2] J. Deisenhofer, D. Braak, H.-A. Krug von Nidda, J. Hemberger, R. M. Eremina, V. A. Ivanshin, A. M. Balbashov, G. Jug, A. Loidl, T. Kimura, and Y. Tokura, Phys. Rev. Lett. **95**, 257202 (2005); J. Q. Li and S. L. Yuan, Solid State Commun. **134**, 295 (2005); H. L. Ibrahim *et al.*, *ibid.* **134**, 695 (2005).

[3] A. Moreo, M. Mayr, A. Feiguin, S. Yunoki, and E. Dagotto, Phys. Rev. Lett. **84**, 5568 (2000); J. Burgy, M. Mayr, V. Martin-Mayor, A. Moreo, and E. Dagotto, *ibid.* **87**, 277202 (2001).

[4] C. Magen, P. A. Algarabel, L. Morellon, J. P. Araújo, C. Ritter, M. R. Ibarra, A. M. Pereira, and J. B. Sousa, Phys. Rev. Lett. **96**, 167201 (2006); Z. W. Ouyang, V. K. Pecharsky, K. A. Gschneidner, Jr., D. L. Schlagel, and T. A. Lograsso, Phys. Rev. B **74**, 094404 (2006); S. Huang, K. Ruan, Z. Lv, H. Wu, Z. Pang, L. Cao, and X. Li, J. Phys.: Condens. Matter **18**, 7135 (2006).

[5] E. V. Sampathkumaran, N. Mohapatra, S. Rayaprol, and K. K. Iyer, Phys. Rev. B **75**, 052412 (2007).

[6] R. B. Griffiths, Phys. Rev. Lett. **23**, 17 (1969).

[7] R. Fisch, Phys. Rev. B **24**, 5410 (1981); V. S. Dotsenko, J. Phys. A **32**, 2949 (1999); A. J. Bray and M. A. Moore, J. Phys. C **15**, L765 (1982); A. J. Bray, Phys. Rev. Lett. **59**, 586 (1987).

[8] L. M. Rodriguez-Martinez and J. P. Attfield, Phys. Rev. B **54**, R15622 (1996).

[9] Wei Li, H. P. Kunkel, X. Z. Zhou, Gwyn Williams, Y. Mukovskii, and D. Shulyatev, Phys. Rev. B **70**, 214413 (2004).

[10] M. Campostrini, M. Hasenbusch, A. Pelissetto, P. Rossi, and E. Vicari, Phys. Rev. B **65**, 144520 (2002).

[11] Wei Li, H. P. Kunkel, X. Z. Zhou, Gwyn Williams, Y. Mukovskii, and D. Shulyatev, Phys. Rev. B **75**, 012406 (2007).

[12] See, for example, D. Kim, B. L. Zink, F. Hellman, and J. M. D. Coey, Phys. Rev. B **65**, 214424 (2002).

[13] A. Arrott and J. E. Noakes, Phys. Rev. Lett. **19**, 786 (1967).

[14] Z. Wang, H. P. Kunkel, and Gwyn Williams, J. Phys.: Condens. Matter **4**, 10385 (1992).

[15] J. H. Zhao, T. Song, H. P. Kunkel, X. Z. Zhou, R. M. Roshko, and Gwyn Williams, J. Phys.: Condens. Matter **12**, 6903 (2000).

[16] J. H. Zhao, H. P. Kunkel, X. Z. Zhou, Gwyn Williams, and M. A. Subramanian, Phys. Rev. Lett. **83**, 219 (1999).

[17] See, for example, N. Rama, M. S. Ramachandra Rao, V. Sankaranarayanan, P. Majewski, S. Gepraegs, M. Opel, and R. Gross, Phys. Rev. B **70**, 224424 (2004).

[18] J. L. Garcia-Munoz, J. Foncuberta, M. Suaaidi, and X. Obradors, J. Phys.: Condens. Matter **8**, L787 (1996).

[19] J. P. Attfield, A. L. Kharlanov, and J. A. McAllister, Nature (London) **394**, 157 (1998).