Critical exponents and the correlation length in the manganite spin glass $Eu_{0.5}Ba_{0.5}MnO_3$

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The critical regime of the half doped manganite spin glass $Eu_{0.5}Ba_{0.5}MnO_3$ is investigated using a combination of the linear and nonlinear magnetic susceptibilities. The divergence of the third-ordered susceptibility (χ_3) signifying the onset of a conventional freezing transition is experimentally demonstrated. The divergence in χ_3, dynamical scaling of the linear susceptibility, and relevant scaling equations are used to determine the critical exponents associated with this freezing transition, the values of which match well with the three-dimensional Ising universality class. Magnetic-field dependence of the spin glass response function is used to estimate the spin-correlation length, which is seen to be larger than the charge and/or orbital correlation length reported in this system.

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I. INTRODUCTION

The hole-doped mixed-valent manganites of the type $AMnO_3$ have been extensively investigated on account of its diverse phase diagram, which in turn arises due to a complex interplay between the lattice, spin, charge, and orbital degrees of freedom. Though earlier emphasis has been on the study of phenomenon like colossal magnetoresistance and charge and/or orbital ordering (CO-OO),[1] more recent investigations have brought forward the phenomenon of *glassiness* in manganites.[2] For instance, large relaxation time scales in measurable quantities such as magnetization, resistivity, and specific heat have been observed in a variety of these systems. In samples of the half doped $La_{0.5}Ca_{0.5}MnO_3$ and $Nd_{0.5}Sr_{0.5}MnO_3$, the magnetization was observed to exhibit long-time logarithmic relaxation rates, with the value of magnetization varying up to time scales of 10^4 s.[3] This slow relaxation was attributed to the coexistence of antiferromagnetic and ferromagnetic (FM) interactions between Mn ions, which in turn produces a distribution of energy barriers. Besides large relaxation times, the phenomenon of *aging*—which refers to the magnetization of a sample cooled in zero field to a certain temperature, being dependent on how long it is held at zero field prior to the magnetic field being applied—has been observed in the system $La_{0.7-x}Y_xCa_{0.3}MnO_3$.[4] The coexistence of FM and non-FM states resulting in novel rejuvenation, persistent memory effects, and dynamical contributions to the magnetotransport have been reported in the system $La_{0.5}Ca_{0.5}Mn_{0.95}Fe_{0.05}O_3$.[5] Nonequilibrium behavior in specific-heat measurements have been observed in $La_{0.5}Ca_{0.5}MnO_3$, which has also been attributed to the dynamics of coexisting phases.[6] Common signatures in bulk magnetization, such as magnetic history effects in dc magnetization and frequency-dependent ac susceptibility, have also been used to identify many manganite systems as having spin glass phases.[7]

Observation of these phenomena, which are normally associated with a conventional spin-glass-like freezing phase transition, has brought to the forefront the issue of whether *glassy* manganites are truly glasses (and thus can be dealt with using conventional spin glass theories) or whether they constitute a fundamentally different state. This issue is further complicated due to the effects of electronic phase separation (EPS), which result in the system subdividing itself into self-organized regions with varying hole concentrations.[8] This EPS could either result in the formation of regions with competing magnetic interactions or in self-generated clusters which can exhibit superparamagneticlike *blocking* at a well-defined temperature.[9] Thus, the fundamental problem here pertains to unambiguously determining whether the observed glassy behavior is purely dynamic (where the average relaxation time of the system exceeds the time scales of the experimental probes) or, on the contrary, is associated with an underlying thermodynamic transition such as in conventional spin glasses.

The prerequisite for a cooperative glassy state is the presence of disorder (either site or bond), which prevents the stabilization of a long-range-ordered magnetic ground state.[10] The same is true within the phase-separation scenario, where disorder promotes the stabilization of self-generated clusters, the interaction between which results in glasslike experimental signatures.[11] Not surprisingly, most manganite systems which exhibit glassy behavior have inherent quenched disorder, either by the direct destabilization of the Mn-O-Mn network through Mn site substitution,[12] or by the random potential which arises from the electrostatic or lattice disorder due to ionic size mismatch between the *A* site ions.[13] The most drastic effect of quenched disorder is seen in narrow bandwidth systems near half doping,[14] which stabilize in the charge-exchange (CE) type of antiferromagnetic ordering which is made up of a checkerboard arrangement of Mn^{3+} and Mn^{4+} ions, accompanied by an associated orbital ordering at the Mn^{3+} sites.[15] The nontrivial geometrical arrangement of the zigzag chains makes it extremely susceptible to disorder, as has been shown by Monte Carlo simulations[14] and experimental investigations of Mn site substituted manganites.[16] It is well known that Mn site disorder results in a destabilization of the long-range magnetic order, leading to magnetic ground states as diverse as relaxor ferromagnets[17] or superparamagnetism.[9] Recently, it has been reported that in narrow bandwidth systems, quenched disorder in the form of large lattice mismatch between the *A* site ions can result in a complete suppression of the CO-OO

state, resulting in a *CE spin glass*.[18] This was observed in half doped samples of the form $A\mathrm{Ba}_{0.5}\mathrm{MnO}_3$. These systems are known to crystallize in two different forms, depending on the synthesis conditions. One variant is the *A* site ordered structure with alternate stacks of *A*O and BaO sheets along the *c* axis with intervening MnO_2 sheets, exhibiting high ($\geqslant$250 K) transition temperatures. The other variant is the *A* site disordered structure (with a disordered distribution of A^{3+} and Ba^{2+} cations), where the random potential originating in the *A* site disorder totally suppresses the long-range order in the charge and orbital sectors, giving rise to a low-temperature ($\leqslant$50 K) spin glass state. Interestingly, this low-temperature state is not associated with macroscopic electronic phase separation, which implies that the spin glass is nearly atomistic,[19] thus making it very attractive as far as investigation of clean manganite glasses are concerned.

The absolute characterization of any phase transition involves the accurate determination of the critical exponents (related to the power-law singularities in various physical parameters), which enable the classification of the phase transition into well-established universality classes[20] which are related to the dimensionality of the lattice and the order parameter. In the context of spin glasses, the appropriate parameter to measure is the nonlinear susceptibility, which by virtue of being an indicator of the Edwards-Anderson order parameter diverges at the freezing temperature.[21] Direct measurement of the nonlinear susceptibility has the added advantage of enabling one to unambiguously distinguish between a spin-glass-like freezing from a superparamagneticlike blocking, since, in superparamagnets, the third-order susceptibility (χ_3) does not diverge and is also known to exhibit a T^{-3} dependence above the blocking temperature.[22] In this paper, we report the investigation of the critical regime of a perovskite manganite glass with an aim to determine the critical exponents and thus the universality class associated with this transition. Since the CE glass is thought to arise from a homogeneous short-range charge and/or orbital order, the spin glass correlation length is estimated in order to verify the validity of the classical Goodenough picture of identical magnetic and orbital correlation lengths.

II. EXPERIMENTAL DETAILS

All the measurements are done on a ceramic sample of $\mathrm{Eu}_{0.5}\mathrm{Ba}_{0.5}\mathrm{MnO}_3$. This sample is prepared using the standard solid-state ceramic route, with starting materials $\mathrm{Eu}_2\mathrm{O}_3$, BaCO_3, and MnO_2 of at least 99.99% purity. Stoichiometric amounts of the starting materials are ground and treated initially at 1000 °C for 12 h, after which they are pelletized and treated at 1500 °C for 36 h with intermediate grindings. X-ray diffraction with a Cu $K\alpha$ laboratory source is used to check the phase purity. The sample is seen to crystallize in a primitive cubic structure (a=3.881 Å) with no observable impurity peaks. The absence of the (0 0 1/2) reflection within the limit of our x-ray diffraction measurement is used to conclude the absence of *A* site ordering as has been done in earlier reports.[23] dc magnetic measurements are done using commercial Quantum Design superconducting quantum interference device and Oxford Maglab vibrating sample magnetometers. Linear and nonlinear ac susceptibilities are measured using a homemade ac susceptometer.

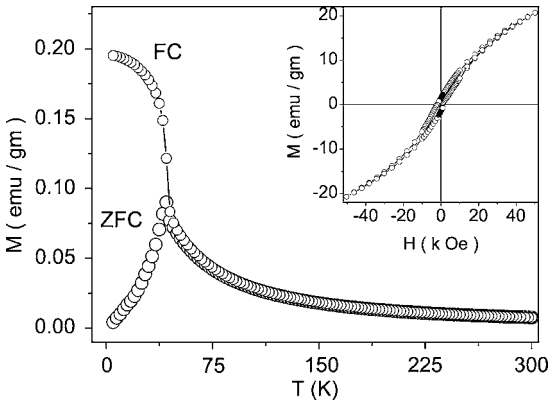

FIG. 1. The bulk dc magnetization as measured in $\mathrm{Eu}_{0.5}\mathrm{Ba}_{0.5}\mathrm{MnO}_3$ in the field-cooled and zero-field-cooled cycles at 100 Oe, exhibiting pronounced irreversibility at the freezing transition. The inset depicts the nonsaturating *M*-*H* isotherm as measured at 5 K.

III. RESULTS AND DISCUSSION

History effects in dc magnetization is a generic feature of nonequilibrium systems such as spin glasses and superparamagnets. Figure 1 shows the magnetic history effect as measured in $\mathrm{Eu}_{0.5}\mathrm{Ba}_{0.5}\mathrm{MnO}_3$, clearly indicating a pronounced irreversibility at $T \approx 42$ K. The inset shows the *M*-*H* isotherm as measured at 5 K, indicating a finite loop width with no trace of saturation up to the highest measured field. Here, it should be noted that in a canonical spin glass, the field-cooled (FC) magnetization is expected to depart from the zero-field-cooled (ZFC) curve at the freezing temperature and remain temperature independent below it. However, a temperature-independent FC behavior is by no means an exclusive feature of spin glasses, as superparamagnets with a narrow volume distribution can also exhibit similar experimental features. It is also well known that in spin glasses and superparamagnets, the presence of a finite dipolar interaction can result in the FC and ZFC curves departing from one another at temperatures higher than the freezing and/or blocking temperatures, with the FC magnetization increasing continuously with decreasing temperature,[24] as is observed in our case.

A more pertinent measurable quantity is the ac susceptibility, which probes the dynamics of the system under investigation at low applied magnetic fields. In spin glasses, the linear susceptibility is expected to show a frequency-dependent cusp (or maximum) at an *effective* spin glass temperature $T_f(\omega)$, with the temperature of the maximum decreasing with decreasing frequency. Figure 2 shows the frequency dependence of the imaginary part of ac susceptibility (χ'') as measured on $\mathrm{Eu}_{0.5}\mathrm{Ba}_{0.5}\mathrm{MnO}_3$, which indicates an enhancement in the peak magnitude as well as a shift in T_f as the frequency of measurement is reduced.

In a continuous magnetic phase transition, when the phase transition temperature is approached from above, the corre-

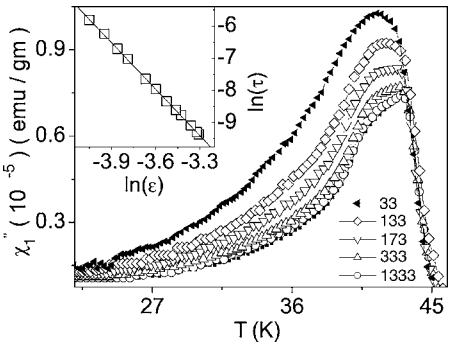

FIG. 2. The frequency dependence of the imaginary part of the linear susceptibility as measured at 3.6 Oe, with the ac frequency being varied from 33 Hz to 1.3 kHz. The inset shows the dynamical scaling of $\tau(T_f)$ with the reduced temperature ϵ, giving T_g=41.05, $z\nu$=7.1±0.2, and $\tau^* \approx 6\times10^{-13}$ s.

lation length (ξ) diverges as $\xi/a=\epsilon^{-\nu}$, where a denotes the average distance between interacting moments, ϵ is the reduced temperature defined as $(T-T_g)/T_g$, with T_g being the *true* spin glass transition temperature, and ν is the critical exponent of the spin correlation length ξ. Assuming conventional critical slowing down on approaching the spin glass transition temperature T_g from the high-T side, the relaxation time τ is related to the correlation length as $\tau \propto (\xi/a)^z$, where z is the dynamic critical exponent. Hence, the T dependence of the relaxation time τ can thus be expressed as

$$\tau = \tau^*(T_f/T_g - 1)^{-z\nu},$$

where τ is $(2\pi f)^{-1}$, with τ^* being the microscopic flipping time of the fluctuating entities. The scaling of τ with the reduced temperature $\epsilon[(T_f-T_g)/T_g]$ is shown in the inset of Fig. 2. In the analysis of τ vs T, we have used values of T_f deduced from the temperature derivatives of $\chi''(T)$. Alternatively, the temperature of the maximum of χ' or χ'' can also be used. The solid line is the fit to experimental data, giving a value of $z\nu$=7.1±0.2, which matches well with that reported earlier on single-crystal specimens.[19] The microscopic flipping time τ^* was found to be 6×10^{-13} s, which is of the same order as that reported for the single-crystal specimens. The true spin glass transition temperature T_g was deduced to be 41.05 K.

By virtue of being proportional to the four spin-correlation functions, the third-ordered susceptibility χ_3 is known to diverge at the spin glass transition and has been used as a signature of the emergence of the Edwards-Anderson order parameter for spin glasses. At the spin glass transition, the third-ordered susceptibility χ_3 exhibits a power-law critical divergence at the spin glass freezing temperature of the form $\chi_3 \propto \epsilon^{-\gamma}$, where the reduced temperature $\epsilon=(T-T_g)/T_g$ and γ is the critical exponent characteristic of a phase transition to a spin glass state. Thus, though the H- and T-dependent peak in the linear susceptibility (χ_1) is non-divergent, the higher-order susceptibility χ_3 diverges in the limits $H\to0$ and $T\to T_g$. This is shown in Fig. 3 using the third-ordered susceptibility (χ_3) data measured using ac susceptibility.[25] The main panel shows a log-log plot of χ_3 as

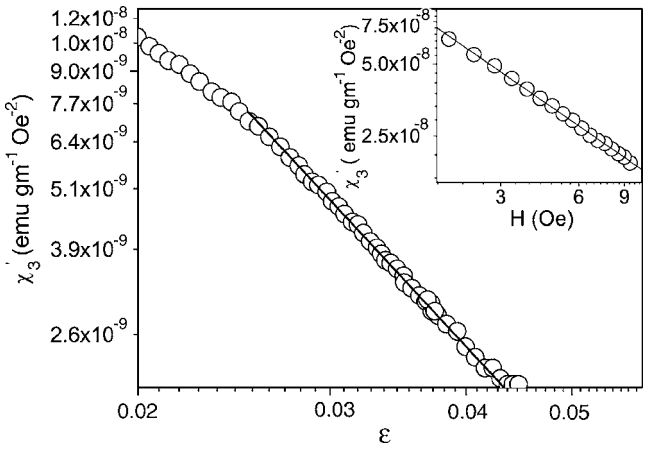

FIG. 3. log-log plot of χ_3 as a function of the reduced temperature (ϵ), indicating the critical divergence of χ_3 in the limit $T\to T_g$. The measurement was done at a frequency of 133 Hz and an ac field of 9.6 Oe, and the straight line fit gives the susceptibility exponent γ=2.74±0.05. The inset shows the log-log plot of χ_3 as a function of the applied field H, indicating the divergence of χ_3 in the limit $H\to0$. The straight line fit gives the value of the field exponent δ, which in this case is deduced to be 4.8±0.5.

a function of the reduced temperature ϵ. The straight fit gives a value of the critical exponent γ=2.74±0.05. The inset shows the log-log plot of χ_3 as a function of the applied field H at the transition temperature T_g. With the order parameter at T_g being proportional to $H^{(2/\delta)}$, the exponent δ can be calculated to be 4.8±0.5 in this case. This report unambiguously demonstrates the critical divergence of the nonlinear susceptibility with H and T in this manganite spin glass, thus confirming that the low-temperature ground state in the system $Eu_{0.5}Ba_{0.5}MnO_3$ indeed arises from a freezing of the spin degrees of freedom.

Dynamical scaling of the ac susceptibility $\chi'(\omega,T)+i\chi''(\omega,T)$ has been extensively used as supporting evidence for critical behavior associated with phase transitions in frozen magnetic systems. A common route is to use the scaling equation

$$\chi'' T/\omega^{\beta/z\nu} \approx g(t/\omega^{1/z\nu}),$$

where g is the scaling function[26] and t is the reduced temperature $(T-T_g)/T_g$. Since the argument of g is now linear in t, a linear scaling plot of $\chi'' T/\omega^{\beta/z\nu}$ resembles χ'' vs T itself, thus enabling one to judge the departure from scaling with relation to the scatter in experimental data. A linear scaling plot of this form, where $\chi'' T/\omega^{\beta/z\nu}$ is plotted against $t/\omega^{1/z\nu}$, is shown in Fig. 4, using the $\chi_1''(\omega,T)$ data as obtained in $Eu_{0.5}Ba_{0.5}MnO_3$. To minimize the probability of erroneous scaling arising from the presence of a large number of free running variables, only the value of β was varied during the course of this scaling procedure. The best scaling was obtained for a value of β=0.6±0.1, with all the peaks in $\chi'' T$ coalescing on the same point.

With the value of the exponents $z\nu$, γ, and β thus obtained, the other exponents can be estimated using the scaling equations

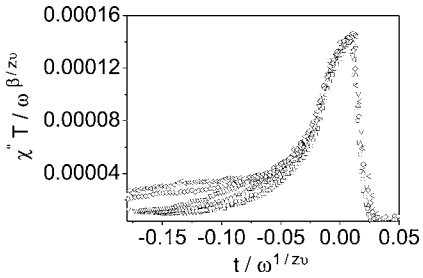

FIG. 4. A linear scaling plot of $\chi'' T/\omega^{\beta/z\nu}$ plotted against $t/\omega^{1/z\nu}$ using $\chi_1''(T)$ data, giving the value of $\beta=0.6\pm0.1$.

$$\alpha + 2\beta + \gamma = 2,$$

$$d\nu = 2 - \alpha,$$

$$\delta = \gamma/\beta + 1,$$

where d refers to the dimensionality and is taken to be 3 in this case. The values of the exponents thus determined is tabulated in Table I. The values of the exponents as expected for a three-dimensional (3D) Ising system from Monte Carlo simulations[27] and those experimentally determined for a well-established 3D Ising system $Fe_{0.5}Mn_{0.5}TiO_3$ (Ref. 28) are also shown for the sake of comparison. As is clearly seen, the values of the exponents determined for the system $Eu_{0.5}Ba_{0.5}MnO_3$ match very well with those expected theoretically and, in some cases, represent an improvement over prior experimental reports, thus unambiguously indicating that the spin glass transition in this manganite system belongs to the 3D Ising universality class.

In this context, it is interesting to note that the exponents measured on the (double-exchange driven) ferromagnetic compositions of various hole-doped manganites have indicated that the double-exchange-driven transitions belong to the isotropic Heisenberg universality class,[29] an observation which is backed up by theoretical predictions that the magnetic interactions are short-range Heisenberg like in such systems.[30] We are not aware of specific theoretical investigations which predict the universality class of low bandwidth manganites like the one investigated in this work. However, one can speculate that the effective anisotropy in our case, which results in an Ising-like exponents, most probably arises as a consequence of short-range orbital ordering which is endemic to narrow bandwidth half doped systems in the presence of disorder. X-ray diffuse scattering measurements indicating the presence of nanometer-scale charge and orbital ordering in $Eu_{0.5}Ba_{0.5}MnO_3$ add credence to this view. Needless to say, the universality class of the system under consideration depends on the dimensionality of the magnetic interactions and the underlying crystal lattice. Recently, a layered manganite spin glass was reported to exhibit exponents which lie between the Heisenberg and Ising universality classes,[31] presumably because the short range orbital order within the two-dimensional layered structure mainly includes $3x^2-r^2/3y^2-r^2$ orbitals favoring in-plane magnetic moments. It is interesting to note that in this layered manganite, the deduced exponents were very close to the Ising universality class and dynamical features such as rejuvenation were seen to be similar to the Ising glasses than the Heisenberg ones.

TABLE I. Critical exponents of the perovskite manganite $Eu_{0.5}Ba_{0.5}MnO_3$ as determined using linear and nonlinear susceptibility data. The values of the exponents as expected theoretically in a 3D Ising system (Ref. 27) and that reported in a well-established Ising system $Fe_{0.5}Mn_{0.5}TiO_3$ (Ref. 28) are also given for the sake of comparison.

Exponent	3D Ising	$Eu_{0.5}Ba_{0.5}MnO_3$	$Fe_{0.5}Mn_{0.5}TiO_3$
γ	2.9±0.3	2.74±0.05	4.0±0.3
β	0.5	0.6±0.1	0.54
z	6.0±0.8	5.4±0.5	6.2
ν	1.3±0.1	1.31±0.08	1.7
α	−1.9±0.3	−1.9±0.25	−3.1
δ	6.8±0.6	4.8±0.5	8.4±1.5

The phenomenon of aging is one of the characteristic features of spin glasses. This occurs due to the fact that the physical property of interest (for example, the magnetization) evolves with time, thus leading to a breakdown of time translational invariance of the response of the system to an external perturbation (for example, the applied magnetic field). In spin glasses, a common avenue of exploring this aspect has been the decay of the thermoremanent magnetization (TRM). In many standard TRM experiments, the spin glass is cooled in the presence of a magnetic field from above the freezing transition temperature (T_g) (i.e., from the paramagnetic phase) down to some measuring temperature T. After waiting for a predetermined time (t_w), the magnetic field is cut off and the decay of the magnetization is measured. This generates the response function, also referred to as the spin glass relaxation rate S, defined as

$$S(t) = d[-M_{trm}(t,t_w)/H]/d \ln t,$$

where $M_{trm}(t,t_w)$ is the thermoremanent magnetization at time t after cutting the magnetic field to zero. $S(t)$ is directly related to the typical value of the free-energy barriers, which can be explored on the available experimental time scales.[32] For any given waiting time (t_w), local equilibration of the state occupancies results in a peak in this response function for measurement times in the vicinity of t_w. The magnetic-field dependence of this peak has been used to estimate the volume over which the spins are effectively locked together for barrier hopping, and thus can be used to estimate the spin glass correlation length.[33]

The M_{trm} as measured in the sample $Eu_{0.5}Ba_{0.5}MnO_3$ is shown in Fig. 5. All measurements were done at $0.7T_f$ after cooling the sample in the presence of a field H from $2.5Tf$. Prior to cutting off the field, the system was made to wait for a time $t_w \approx 60$ s. The inset shows the magnetic-field dependence of the response function $S(t)$, clearly indicating that the measuring time at which it peaks decreases as a function of the magnetic field H. This change in the response function has been explained using models, where the phenomenon of

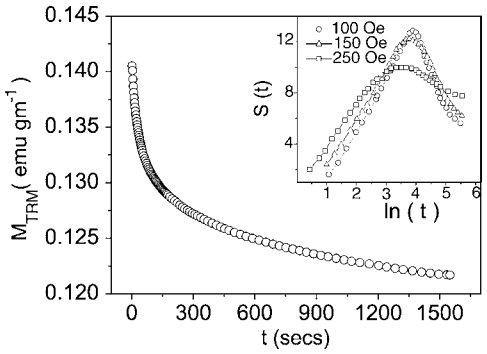

FIG. 5. M_{TRM} as measured at $0.7T_f$ after cooling in the presence of a field of 100 Oe and waiting (t_w) for 60 s. The inset shows the magnetic-field dependence of the relaxation rate $S(t)$ at applied fields of 100, 150, and 200 Oe, which is used to estimate the correlation length ξ_{mag}.

aging in spin glasses have been treated as a random walk within a wide distribution of traps.[32] When the magnetic field is changed, all traps have their depth reduced by a Zeeman energy term (E_z). This change in the barrier heights results in an effective *reduction* of the measuring time at which $S(t)$ peaks. This peak can then be associated with an effective waiting time t_{eff}. This apparent age t_{eff} (smaller than the actual waiting time t_w) is given by $\ln t_{eff}/t_w = -E_z/k_BT$ when the change in $S(t)$ is associated with a magnetic energy E_z. As was done before for Ising systems,[34] this Zeeman energy term E_z can be equated to $\sqrt{NH}(m\mu_B)$, where N is the number of spins which are effectively blocked together and $m\mu_B$ is the effective moment of one spin entity.

The value of N thus calculated in this system is seen to be of the order of 10^5,[35] which is larger than that reported for the Ising system $Fe_{0.5}Mn_{0.5}TiO_3$ and comparable with that reported in Heisenberg glasses.[34] The effective correlation length (ξ_{mag}) can then be estimated to be of the order of 35 Å,[36] which is larger than the charge-orbital correlation length (ξ_{orb}) estimated for this system[18] by a factor of 1.75. However, it should be borne in mind that the earlier estimation of ξ_{orb} was made using the width of the x-ray diffuse scattering, a technique which provides a more direct and reliable value of the correlation length. Interestingly, soft x-ray resonant experiments on a COO manganite near half doping have shown that the magnetic correlation length exceeds that of the orbital order by a factor of 2.[37] This challenges the classical Goodenough model, where orbital domain walls create magnetic domain walls in the Mn^{3+} sublattice which, in turn, would imply that the ratio of ξ_{mag}/ξ_{orb} is unity. If exact, our estimates of ξ_{mag} would indicate that a ratio of $\xi_{mag}/\xi_{orb} \approx 2$ is valid not only for OO systems with concomitant long-range magnetic order, but also in systems where disorder results in a suppression of magnetic long-range ordering. Needless to say, more accurate magnetic scattering experiments would be needed to corroborate this conjecture. The temperature (T^*),[38] where these short-range correlations in this system would form is $\approx 2.5T_f$ as can be estimated from the deviation from linearity of χ^{-1} in the paramagnetic regime.

IV. CONCLUSIONS

In summary, we have investigated the critical regime of the CE spin glass $Eu_{0.5}Ba_{0.5}MnO_3$ using linear and nonlinear ac susceptibilities. The critical divergence of the third-ordered susceptibility (χ_3) as a function of H and T is demonstrated, thus confirming that the low-temperature metastable magnetic phase arises from the freezing of the spin degrees of freedom. The exponents determined unambiguously show that this transition falls into the 3D Ising universality class. The spin glass correlation length is estimated from magnetic-field change aging experiments and indicates that this magnetic correlation length is larger than the charge and/or orbital length reported earlier. Keeping in mind the fact that recent experiments have challenged the basic premises of the classical Goodenough model of CE systems, our results call for more systematic investigations of the charge and/or orbital and magnetic length scales in such systems.

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