Cluster-glass dynamics of the Griffiths phase in $Tb_{5-x}La_xSi_2Ge_2$

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The static magnetization and dynamic susceptibility responses of the cluster system within a Griffiths phase of the magnetocaloric compound $Tb_{5-x}La_xSi_2Ge_2$ ($x = 0.075$) have been investigated. A novel cluster-glass state within the Griffiths phase is formed at a characteristic freezing temperature where short-range ferromagnetic correlations set in the paramagnetic regime. Ferromagneticlike correlations are built up at around 155 K, which suddenly become frozen at a lower temperature ~140 K, thus in analogy with a reentrant spin glass behavior. The ac susceptibility near the freezing temperature follows a critical slowing down process characterized by $\tau_0 = 10^{-13}s$ and dynamic exponents $z\nu \sim 6$ and $\beta \sim 0.4$, similar to well-known spin glass systems. The nonlinear ac susceptibility analysis shows clearly the existence of a transition associated to the reentrant behavior. The origin of the intermediate cluster-glass phase inside the Griffiths phase is proposed to be the result of a combination of short-ranged RKKY intralayer positive exchange interactions between rare-earth Tb^{3+} ions and antiferromagnetic exchange between adjacent interlayers involving Si and Ge atoms in connection to the Tb^{3+} atoms.

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I. INTRODUCTION

The $R_5(Si_xGe_{1-x})_4$ (R = rare earth) compounds have been a subject of intensive research since the discovery of the giant magnetocaloric effect (GMCE) near the magnetostructural transition in $Gd_5Si_2Ge_2$ compound [1,2]. Their promising magnetoresponsive properties, e.g., giant magnetocaloric effect (MCE) [1], giant magnetoresistance [3] and colossal magnetostrictive effect [4], have triggered a plethora of studies in order to shed light on the nature of such intense effects. The physical properties of the $R_5(Si_xGe_{1-x})_4$ system are determined by a special crystallographic structure involving atomic layers forming rigid and compact slabs of R and T = Si/Ge atoms [5] where tiny modifications can modify greatly the magnetic coupling due to the competing ferromagnetic (FM) and antiferromagnetic (AFM) interactions.

The crystallographic phase and magnetic ordering are controlled by the number of interlayer covalentlike T-T bonds connecting the slabs [6]. The rearrangement of these slabs via changing specific T-T bonds gives rise to the three main crystal structures adopted by this family of materials. When all the T-T pairs are covalently bonded the system adopts the Gd_5Si_4 structure type [also referred to as $O(I)$ state with a space group *Pnma*], whereas those without any interslab T-T bonding possess the Sm_5Ge_4-type structure [referred to as $O(II)$ structure, space group *Pnma*]. When half of the bonds are broken, this system presents a $Gd_5Si_2Ge_2$-type [M state, space group $P112_1/a$].

From the magnetic standpoint, two types of magnetic interactions are present in these alloys: the intralayer interaction (J_{intra}) and the interlayer interactions (J_{inter}), via the existing T-T bonds. Both interactions are believed to be ruled by the conventional $4f$-$4f$ Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect exchange [7]. Therefore, the crystallographic structure and the magnetic properties in $R_5(Si_xGe_{1-x})_4$ compounds are closely related since both are controlled by the number of the interlayer T-T bonds connecting the slabs [6]. Both theoretical and experimental work have been reported supporting the picture that the intralayer magnetic structure is essentially ferromagnetic (FM), whereas the sign of J_{inter} depends on the distance between the slabs and tends to be either FM or antiferromagnetic (AFM), depending on the number of interslab pairs that are covalently bonded [8,9]. Thus, J_{inter} displays a FM character in the case of monoclinic (M) and orthorhombic $O(I)$ structures (where the slabs are partially or totally bonded), or AFM for the orthorhombic $O(II)$ structure (when the slabs are unbonded).

The complex layered crystalline structure, the competition between magnetic interactions, and the strong relation between structure and magnetic properties in this system lead to the observation of another exotic regime: the Griffiths

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phase (GP). According to Griffiths [10], a randomly diluted FM will exhibit a suppressed long-range ordering temperature $T_C(x)$ (where x is the dilution), and in the temperature region $T_C(x) < T < T_C^{\text{undiluted}}$(where $T_C^{\text{undiluted}}$ is the T_C of the nondiluted FM) the thermodynamic properties will become nonanalytical due to the formation of a low density of short-range ordered clusters. $T_C^{\text{undiluted}}$ is therefore the temperature at which the GP forms and has been labeled as Griffiths temperature T_G. The small clusters give rise to characteristic features that allow the identification of the GP, namely, the deviation of the reciprocal susceptibility (χ_{dc}^{-1}) from the Curie-Weiss predictions as the system approaches T_C on cooling from $T < T_G$, taking the form of an enhanced low field susceptibility [11]. Such a deviation is suppressed by a moderate magnetic field (H_{dc}) due to the polarization of spins outside the clusters [12,13].

The onset of GP within this family of compounds was first reported in $Tb_5Si_2Ge_2$ [13] by means of dc magnetic susceptibility and small-angle neutron scattering (SANS) experiments. Subsequent work by Pereira *et al.* [14] also revealed the presence of short-range magnetic correlations in the monoclinic (M) and orthorhombic $O(II)$ structural phases for the $R_5(Si_xGe_{1-x})_4$ system with $R =$ Gd, Tb, Dy, and Ho. According to these studies, T_G was identified as the T_C of the pure FM phase ($T_C^{\text{undiluted}}$) which in this case has an orthorhombic $O(I)$ structure. Therefore, the FM clusters responsible for the GP signatures in the experimental data were proposed to be due to nanometric regions of orthorhombic character $O(I)$ and strengthened FM interactions.

Chemical substitution has been used as a tool to stabilize atomic and magnetic structures and, consequently, to tune the magnetoresponsive properties of the $R_5(Si_xGe_{1-x})_4$ system. Particularly one can attempt the substitution of the rare-earth $R_{1-x}R'_x(Si,Ge)_4$; several studies have been carried out in Gd-based compounds [15,16]. In this context, Mudryk and coworkers were able to manipulate individually the R atoms, using nonmagnetic R such as La and Lu, which tune the magnetic properties of the parent compound Gd_5Ge_4 [17]. They observed that despite the substitution of a magnetic ion (Gd) by small concentrations of a nonmagnetic ion (La), the magnetic behavior of the compound did not change significantly, since such a low substitution did not interfere with the atomic chain responsible for the interslab ferromagnetic interactions (R_1-T_3-T_3-R_1). However, a similar amount of Lu substitution was shown to alter drastically the magnetic properties of the parent compound. Such a behavior was explained by the preferential Lu occupation of R_1 sites, which interfere with exchange interactions of the R_1-T_3-T_3-R_1 chains. Inspired by these results, Belo and coworkers extended the study to $Tb_5Si_2Ge_2$, in order to understand the role of nonmagnetic atoms (La) on parent compounds exhibiting spontaneous magnetostructural transitions and on how it could affect the magnetic exchange mechanism [18]. In that sense, $Tb_5Si_2Ge_2$ is an ideal system to carry out such a study since it presents two distinct phase transitions separated by approximately 10 K: one purely magnetic [PM, M] $\rightarrow$ [FM, M], followed by a structural change [FM, M] $\rightarrow$ [FM, $O(I)$] at lower temperatures [19].

By means of a thorough study of the structural and magnetic properties of the $Tb_{5-x}La_xSi_2Ge_2$ system, they found

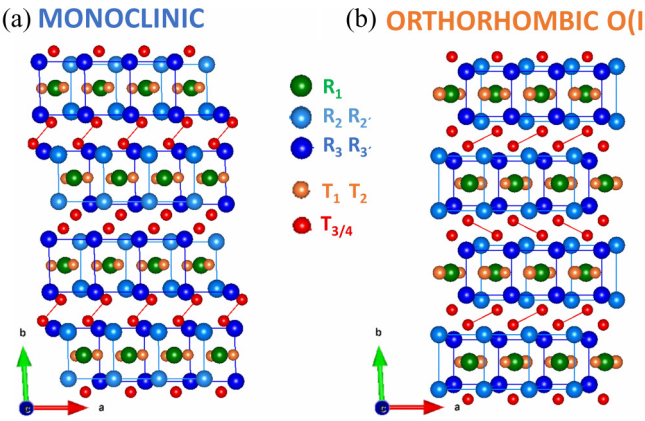

FIG. 1. Crystal structures of $Tb_5(Si_2Ge_2)$ in the paramagnetic (a) and in the ferromagnetic state (b). The R atoms occupying different sites are shown using green (R_1), light blue (R_2), and dark blue (R_3) spheres. The orange circles represent the Si/Ge atoms located at the T_1, T_2 positions within the slab. The red circles represent the Si/Ge atoms responsible for bonding between slabs, located at the T_3, T_4 positions. The thick red lines indicate the Si/Ge-Si/Ge covalently bonded pairs of atoms.

that La ions influenced the relation between atomic and magnetic structure by fully coupling the magnetic and structural transitions from [PM, M] to [FM, $O(I)$] for small La concentrations ($0.25 < x < 1$). Figure 1 shows the crystallographic structure of these compounds in the PM state (M structure) and in the FM state ($O(I)$ structure). Despite the substitution of a magnetic (Tb) for a nonmagnetic ion (La), an increase of T_C with La concentration was observed in the $x < 1$ regime. This feature was also explained by the preferential La occupation of the R_2 site [R_2 site in the $O(I)$ structure is an identical site to the R_2 site in the M structure], which was supported by density functional theory calculations. Thus, La atoms do not interfere with the interslab FM interaction at the R_1-T_3-T_3-R_1 chain [18]. The increase in T_C is due to the fact that the R_2 site is the one exhibiting higher spin canting in the parent compound $Tb_5Si_2Ge_2$ [20,21] and thus, the La occupation of this site favors the collinearity of the Tb magnetic moments (i.e., a decrease of AFM interactions between Tb atoms). The La occupation of the R_2 site also explains why the magnetic behavior of the parent compound did not change significantly for small La concentrations ($x < 0.25$).

In this paper we experimentally show that a small amount of La ($x = 0.075$) is able to affect the mechanisms responsible for the formation of the short-range FM clusters in the GP in the parent compound, $Tb_5Si_2Ge_2$. For this, we have selected key techniques to analyze the static behavior (dc magnetization) and the magnetic relaxation of the clusters (linear and nonlinear ac susceptibility) in the Griffiths phase ($T > T_C$) of $x = 0$ and $x = 0.075$ compounds. These measurements provide significant information about the spin dynamics in this clustered system, due to the intrinsic sensitivity of ac susceptibility (related to its inherent derivative nature) to detect very subtle magnetic transitions and/or different magnetic phases. Moreover, the simultaneous detection of nonlinear magnetic ac susceptibility is able to unambiguously distinguish between

complex clustered systems and directly probe the magnetic order prevalent within the clusters [22].

II. EXPERIMENTAL DETAILS

The polycrystalline $Tb_{5-x}La_xSi_2Ge_2$ samples with $x = 0$ and $x = 0.075$ were prepared using an arc-melting furnace. The starting elements were 99.99 wt% pure Tb, 99.9 wt% pure La, and 99.9999 wt% pure Si and Ge (Alfa Aesar). Further details on sample preparation and basic structural characterization at room temperature can be found elsewhere [13,18]. The dc magnetization measurements were performed in a commercial (Quantum Design) superconducting quantum interference device (SQUID) magnetometer in applied dc magnetic fields from $H_{dc} = 0.02$ kOe up to $H_{dc} = 1$ kOe in the temperature range 2–300 K. The ac susceptibility measurements were carried out in a commercial physical property measurement system (PPMS) with the AC measurement system (ACMS) option. Both real $\chi'_{ac}(T)$ and imaginary $\chi''_{ac}(T)$ components of the linear susceptibility were recorded using an ac magnetic field $h_{ac} = 1$ Oe in amplitude: (a) at several biasing dc magnetic fields in the range from $H_{dc} = 0$ to 1 kOe with ac frequency $f = 100$ Hz and (b) at several frequencies f from 100 to 10 000 Hz with $H_{dc} = 0$. To extract the coefficients of the nonlinear susceptibility, it is necessary to detect with a lock-in amplifier the voltage (which is frequency dependent) generated in the pick-up coils. The use of the lock-in signal allows us to extract directly and simultaneously the signal harmonics. The utility of these harmonics will be explained in more detail in the next section.

III. RESULTS AND DISCUSSION

Figure 2(a) shows the magnetization data in field-cooled (FC) and zero field-cooled (ZFC) protocols for $x = 0.075$ at an applied field $H_{dc} = 0.02$ kOe. In the FC protocol, the sample is cooled to 5 K in the presence of the measuring field and the magnetization is recorded in a heating run keeping the field constant. In the ZFC protocol the sample is cooled to 5 K in zero field and then the measuring field is applied and the magnetization is recorded as a function of temperature in the heating run. The Curie temperature occurs at $T_C = 105(1)$ K (obtained by the minimum of the numerical derivative dM/dT) and the second anomaly observed at $T_{SR} = 65$ K is assigned to a spin reorientation process similar to that observed in the parent compound $Tb_5Si_2Ge_2$ [20,21]. The irreversibility observed between the ZFC and FC magnetization data below T_C reflects sizable pinning effects in this FM compound. It is remarkable the appearance of irreversibility well above T_C, which is manifest as a bifurcation between the ZFC and FC curves below a characteristic temperature $T_{ir} \sim 160$ K.

The inverse of low field dc susceptibility χ_{dc}^{-1} of $x = 0.075$ compound is shown in the inset of Fig. 2(a). On cooling, χ_{dc}^{-1} follows the typical Curie-Weiss behavior in the high-temperature region that however disappears at $T_G \sim 190(1)$ K above the ordering temperature T_C. Such a deviation indicates the appearance of short-range magnetic clusters in the paramagnetic (PM) state and represents the fingerprint of a Griffiths phase, as observed in the parent compound $Tb_5Si_2Ge_2$ [13].

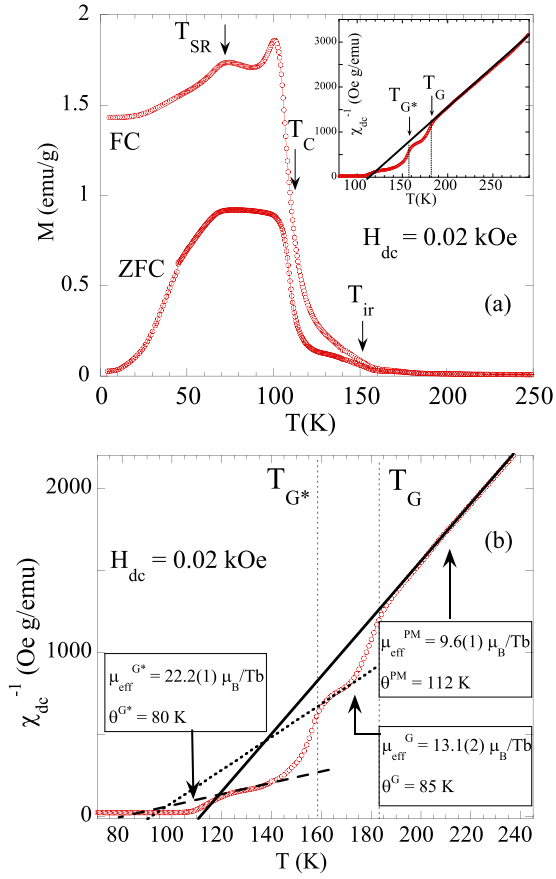

FIG. 2. (a) ZFC-FC magnetization of $Tb_{4.925}La_{0.075}Si_2Ge_2$ as a function of temperature measured in an applied field of 0.02 kOe. The inset of Fig. 2(a) shows the Curie-Weiss fit to the inverse magnetic susceptibility in 0.02 kOe. (b) Temperature dependence of dc magnetic susceptibility of $b_{4.925}La_{0.075}Si_2Ge_2$ plotted as χ_{dc}^{-1} (T) in the temperature range 70–240 K, measured in a field of 0.02 kOe. Curie-Weiss fit is shown for the pure PM region (solid line) and for the temperature regimes delimited by T_G and T_{G^*} (dashed lines).

In the present case, an additional stairlike fall in χ_{dc}^{-1} is seen below a characteristic temperature $T_{G^*} \sim 156(1)$ K. This steplike anomaly defines two different plateaus in χ_{dc}^{-1} in the temperature ranges 160–180 K and 120–140 K, respectively. The different slopes observed in χ_{dc}^{-1} in both temperature regimes indicate different sizes of the FM clusters in these temperature regimes. Such a stairlike behavior in χ_{dc}^{-1} was also observed in the parent compound, $x = 0$ [13]. In that case, however, the low temperature anomaly in χ_{dc}^{-1} was much weaker.

The fit of χ_{dc}^{-1} to the Curie-Weiss law [Fig. 2(b)] provides the effective magnetic moment μ_{eff} and the extrapolated paramagnetic Curie temperatures θ_P associated with each of the different temperature regimes. The value of μ_{eff} in the PM regime ($T > 190$ K) is $\mu_{\text{eff}}^{PM} = 9.6\ (1)\ \mu_B$/Tb, which is close to the expected value for free Tb^{3+} ions ($\mu_{\text{eff}}^{PM} = 9.72\ \mu_B$/Tb). The same fit in the 160–174 K plateau yields a larger value ($\mu_{\text{eff}}^{G} = 13.1(2)\ \mu_B$/Tb) indicating the FM clustering of spins below T_G, and the value rises up to $\mu_{\text{eff}}^{G^*} = 22.3(2)\ \mu_B$/Tb in the lower-temperature plateau (below T_{G^*}), as the true long-range ferromagnetic ordered state is approached. The

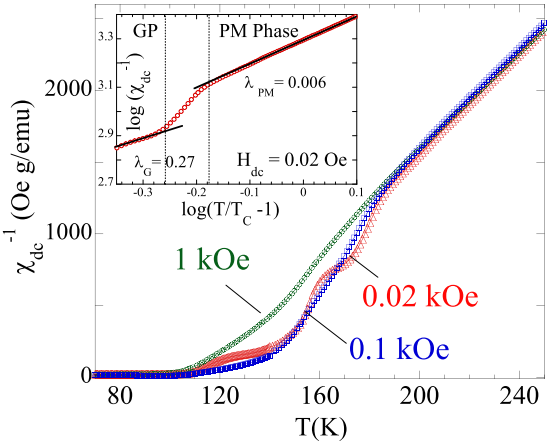

FIG. 3. Temperature dependence of dc magnetic susceptibility χ_{dc}^{-1} as a function of magnetic field, measured on heating. The inset shows fits at 0.02 kOe in the Griffiths (GP) and PM phases, respectively.

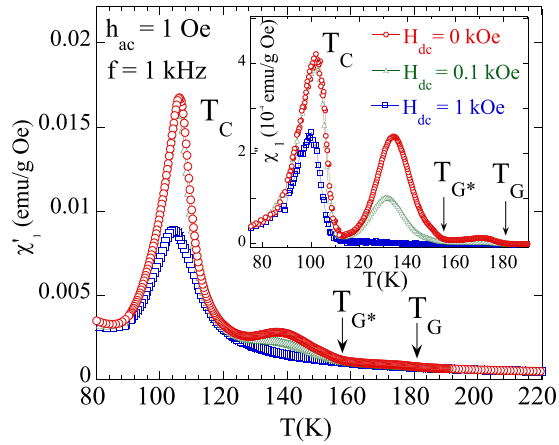

FIG. 4. Real part of the ac-susceptibility χ_1' measured at different applied dc fields (0, 0.1, and 1 kOe). The inset shows the imaginary part of the ac susceptibility χ_1'' in the temperature range 75–190 K measured at the same applied dc fields.

same analysis in the parent compound $x = 0$ yields similar values ($\mu_{\text{eff}}^{G} = 13\mu_B$/Tb and $\mu_{\text{eff}}^{G^*} = 23\ \mu_B$/Tb) [23]. Such a similarity indicates that both compounds have FM clusters of similar sizes in the PM state. Overall, those results indicate that the different plateaus in χ_{dc}^{-1} depict different stages of the FM clustering of spins within the GP.

Figure 3 shows that both stairlike jumps in χ_{dc}^{-1} for $x = 0.075$ compound are affected by the dc field: On further increasing the magnetic field up to $H_{dc} = 0.1$ kOe, the steplike anomaly at 156 K in χ_{dc}^{-1} vanishes, and thus, the lower temperature plateau is suppressed. When the applied field is $H_{dc} = 1$ kOe no trace of the steplike behavior is observed and χ_{dc}^{-1} becomes nearly indistinguishable from the high-temperature range values, within the experimental error. This indicates that the cluster FM component is weak and masked at relative low fields by the linear increase of the PM contribution of the matrix, as already observed in the parent compound $Tb_5Si_2Ge_2$ [13].

It is known that the Griffiths phase is characterized by a power law behavior of the inverse of the susceptibility [24] $\chi_{dc}^{-1}(T) = (T/T_C - 1)^{1-\lambda}$, where $0 < \lambda < 1$, in the intermediate region $T_C < T < T_G$, and close to zero $\lambda_{PM} \approx 0$ in the PM regime. We have fitted the logarithmic representation of χ_{dc}^{-1} obtaining clearly different values for the exponent λ depending on whether we analyze the anomalous region of χ_{dc}^{-1} or in the conventional paramagnetic phase (see inset of Fig. 3). It is worth mentioning that, in this study, the fit of the low field χ_{dc}^{-1} must be done in the temperature regime that defines the first plateau 160 K $< T < T_G$ to avoid the contribution of the lower-temperature anomaly in χ_{dc}^{-1}. The corresponding values of the exponent in the so-defined anomalous region [$\lambda_G = 0.27(2)$ at 20 Oe] remains far from zero in this case, whereas it remains very small [$\lambda_{PM} = 0.006(1) \approx 0$ at 20 Oe] in the conventional paramagnetic regime ($T > 200$ K). At a higher field ($H_{dc} = 100$ Oe), when the lower temperature anomaly in χ_{dc}^{-1} is suppressed, the linear fit of χ_{dc}^{-1} can be extended down to $\sim T_C$ (118 K $< T < T_G$). The corresponding values of the exponent remain far from zero value also in this case, e.g., $\lambda_G = 0.20(1)$ at 0.1 kOe, so the Griffiths state remains stable. In contrast, λ_{PM} becomes very small [$\lambda_{PM} = 0.001(1)$ at 0.1 kOe], indicating that the Griffiths phase does not extend to temperatures higher than 190 K. It is worth mentioning that GP remains even at higher fields (1 kOe), where $\lambda_G = 0.13(1)$ and $\lambda_{PM} = 0.004(1)$. This observation supports that the intermediate clustered environment does not disappear with the applied magnetic field and the magnetic field suppression of the anomaly displayed by χ_{dc}^{-1} is explained in terms of the masking of the short-range FM signal by the rising PM background.

Interestingly, the real [χ_1' (T)] and imaginary [χ_1''(T)] components of the ac magnetic susceptibility as a function of temperature (Fig. 4) recorded at a single frequency $f = 1000$ Hz without dc magnetic field, evidence two broad peaks centered at $T = 172.0(5)$ K and $T = 136.0(5)$ K. The fact that the anomalies appear simultaneously in χ_1' (T) and χ_1''(T) indicates that both transitions are accompanied by an energy dissipation process, usually associated with domain dynamics. Both peaks in χ_1'(T) and χ_1''(T) are affected by the biasing (superposed) small dc field: When increasing H_{dc} both peaks gradually diminish being barely observable for $H_{dc} = 1$ kOe. Such a magnitude of the bias field, however, is not sufficiently high to suppress the anomaly in χ_1''(T) associated to T_C. Those evidences indicate the large influence of the biasing field on the dynamic response of the clusters, which may be connected to a situation in which the ferromagnetic order is not well established between T_G and T_{G^*} [25].

The unexpected anomaly observed within the GP at $T = 136$ K could be associated to the magnetic ordering temperature of a La-richer $Tb_{5-x}La_xSi_2Ge_2$ impurity phase (e.g., $T_C = 140$ K for $x = 0.5$) [18]. However, this possibility must be ruled out bearing in mind the following evidences. The x-ray powder diffraction refinement analysis revealed a negligible amount of secondary phase with the *R*: (Si,Ge) = 1:1 stoichiometry (a volume fraction of less than 4%) whereas no traces of a phase with higher La content were detected (within the sensitivity of the technique, which can be estimated as 2–5 vol.% of an impurity phase) [18]. It is worth recalling

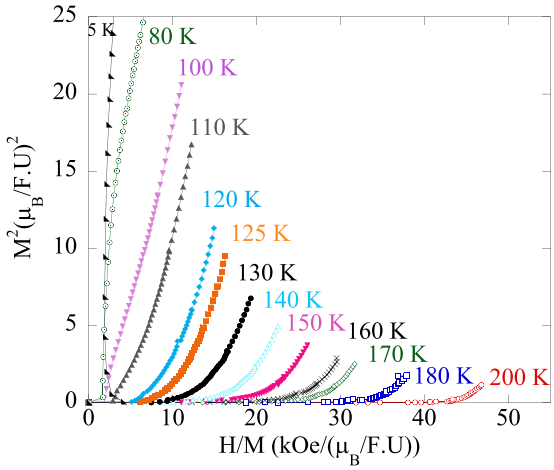

FIG. 5. Arrott plots (M^2 vs H/M) of the magnetization isotherms at selected temperatures in the temperature range 5–200 K.

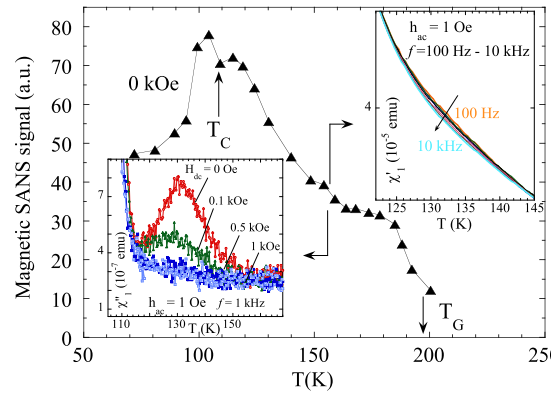

FIG. 6. Temperature dependence of the magnetic SANS intensity for $Tb_5Si_2Ge_2$ collected in D16 at $q = 0.1$ Å^{-1} at zero magnetic field from Magén *et al.* [13]. The lower inset shows the temperature variation of the imaginary component of the ac susceptibility (χ_1'') in the temperature regime around 150 K, where the plateau in SANS was observed, collected under various dc bias fields (H_{dc}). The upper inset shows the temperature variation of the real component of the ac susceptibility (χ_1') in the same temperature range at different frequencies (f = 100, 215, 463, 1000, 2150, 4630, and 10 000 Hz).

that the 1:1 phases order antiferromagnetically below ∼60 K [26], which is far from the observed anomaly at 136 K. To further inspect the possibility of magnetic ordering due to a secondary phase, Arrott plots (M^2 vs H/M) were performed and are shown in Fig. 5. In this kind of plot, the intercept of the linear fitting of high field data on the Y axis provides the spontaneous magnetization and the fit passing through the origin gives the ferromagnetic transition temperature T_C. In this case, the line passing through origin corresponds to the M^2 versus H/M curve lying in between 100 K and 110 K indicating that the T_C lies in between. Above T_C, the spontaneous magnetization is zero, indicating the absence of long-range ferromagnetic ordering and thus excluding the possibility of existence of a secondary phase.

It is worth noting that a similar feature in the GP regime is observed also in the parent compound $Tb_5Si_2Ge_2$. Insets of Fig. 6 show χ_1'(T) recorded at several frequencies and χ_1''(T) recorded at a single frequency $f = 1000$ Hz at several dc magnetic fields as a function of temperature in the GP temperature regime. In addition to the maximum corresponding to the Curie temperature $T_C = 101.5(5)$ K and the tiny kink at $T_G = 200.0(5)$ K (not shown), a peak appears in χ_1''(T) at $T = 131.2(3)$ K. This peak is also affected by the biasing dc field, being totally suppressed with a $H_{dc} = 1$ kOe. Compared with the $x = 0.075$ compound, this peak is smaller in magnitude. Moreover, this anomaly is not distinguished in χ_1'(T) although a frequency dependent signal is observed around this temperature. Furthermore, such a peak in χ_1''(T) appears in the temperature regime where an anomalous plateau of the SANS signal was reported below T_G (published data now inserted in main panel of Fig. 6) [13]. Such an anomalous contribution was associated with the nucleation of FM clusters within the PM region of $Tb_5Si_2Ge_2$. According to the observed behavior in χ_1''(T), there is a progressive freezing of the clusters forming at T_G which can be understood as a relaxation process of the moments within the magnetic clusters to finally become randomly oriented (frozen). This frozen state of the Griffiths-like clusters is achieved above the long-range ferromagnetic ordering temperature T_C.

The static and dynamic susceptibility results indicate that a (weak) magnetic state is created within the GP (below T_G) in both compounds. In order to reveal the dynamic nature of this state we have carried out a deeper study of the ac susceptibility in the $x = 0.075$ compound, since the anomaly is more pronounced than in $x = 0$. Figure 7 shows the temperature dependence of the real (χ_1') and imaginary (χ_1'') components of the ac magnetic susceptibility of $x = 0.075$ without dc magnetic field measured at various frequencies (f in the range of 100–10000 Hz). Both χ_1' and χ_1'' go through a broad peak, centered around $T \sim 136$ K at $f =$ 100 Hz. The position of the χ_1' maximum shifts to higher temperature and the amplitude decreases with increasing ac driving-field frequency f. We observe that the peak shifts up to ∼140 K when the frequency varies up to 10000 Hz. Clearly, the spin dynamics of this transition (revealed by the peak) is essentially similar to that of a disordered magnet, i.e., a spin glass. In this sense, the observation that the peak position depends on frequency precludes the possibility of a true thermodynamic FM to PM phase transition but instead reflects the behavior normally associated with spin glasses. If the peak temperature is identified with the spin-glass freezing temperature T_f, we can estimate the relative variation in T_f per decade of frequency $f(\delta)$:

$$\delta = \frac{\Delta T_f}{T_f \Delta(\log_{10} f)} \tag{1}$$

which is a quantitative measure of the effect of the frequency in shifting the peak of χ_1'(T). The δ value allows the comparison among the f dependence of the freezing temperature T_f in different systems. For $x = 0.075$ we found $\delta = 0.01$, which is an intermediate value between those reported for canonical spin-glass systems ($\delta = 0.005$ for CuMn) [27] and those reported for noninteracting ideal superparamagnetic systems ($\delta = 0.1$) [28]. Thus, the value of $\delta = 0.01$ best corresponds to an intermediate situation, the so-called cluster glass. Exam-

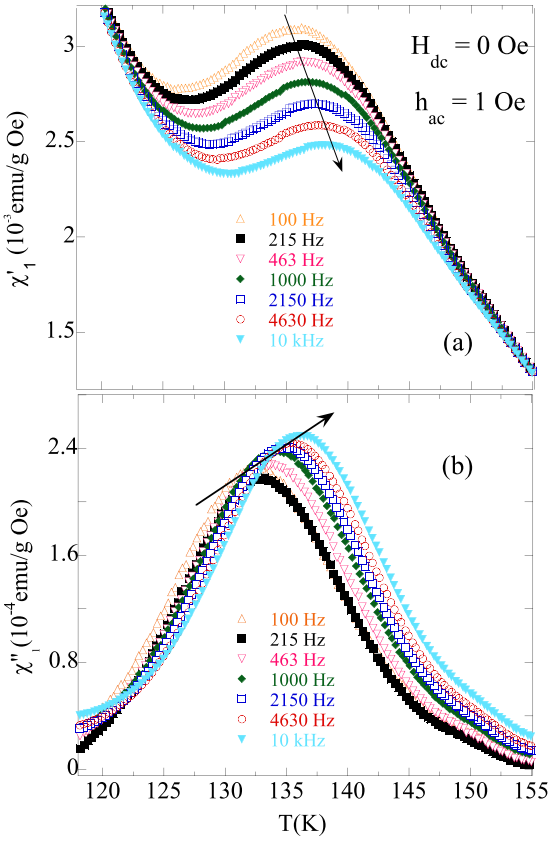

FIG. 7. The real (χ_1') and imaginary parts (χ_1'') of the ac susceptibility as a function of temperature of $Tb_{4.925}La_{0.075}Si_2Ge_2$ measured at different frequencies from 100 Hz to 10 kHz in an applied ac field of $h_{ac} = 1$ Oe in the temperature range 115–155 K.

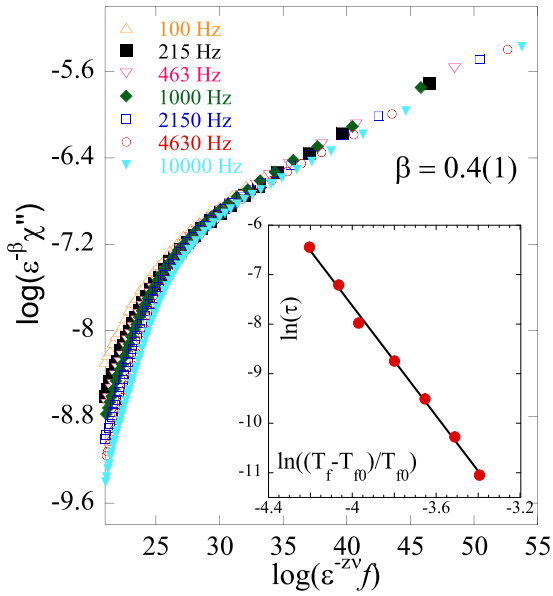

FIG. 8. Critical slowing down analysis of the imaginary part of the ac magnetic susceptibility data χ_1'' at $T > T_f$ [see Eqs. (3) and (4)] of the $x = 0.075$ alloy. χ_1'' at different frequencies of the ac field collapse on a master curve with $\beta = 0.4(1)$. The inset shows the frequency dependence of freezing temperature T_f plotted as a $\ln(\tau) vs \ln[(T_f\text{-}T_{f_0})/T_{f_0}]$ (see text for details). The solid line represents the fit to the power-law divergence of critical slowing down process Eq. (3).

ples of δ values of this magnitude can be found in other metallic systems such as $Pr_{0.8}Ca_{0.2}MnO_3$ [29], $CeNi_{1-x}Cu_x$ [30], and in the Heusler alloy Ga_2MnCo [31]. Another important observation is that the χ_1'(T) peak is much broader and χ_1'(T) is one order of magnitude larger than that in the canonical spin glasses [32,33]. A broad peak and the enhancement in χ_1'(T) are indicative of the presence of FM clusters. It is worth recalling that for the case of a second order transition with a well-defined ordering temperature T_C (FM) or T_N (AFM), frequencies ranging from megahertz to gigahertz are required to detect a shift in the maxima [27]. This fact together with the absence of spontaneous magnetization in the Arrott plots shown above clearly rule out the possibility of a long-range magnetically ordered state at 136 K.

The combination of such an intermediate δ value, a relatively broad relaxation, and the susceptibility value supports clearly the presence of magnetic clusters (Griffiths origin) in which interactions are present. In such a case, the dynamics may be better accounted by a critical slowing down process as the system approaches the phase transition. To analyze the critical behavior in this process is necessary to measure the divergence of the relaxation time τ as the temperature approaches the critical freezing temperature T_{f_0} at which the phase transition takes place. Here, T_{f_0} represents the infinitely slow cooling dc (equilibrium) value of T_f (i.e., peak in χ_1' when $f \to 0$). The conventional result of dynamical scaling relates the relaxation time τ due to correlated dynamics to the spin-spin correlation length ξ as $\tau \sim \xi^z$, z being the dynamic critical exponent. Since ξ diverges with temperature as $\xi \sim [T/(T\text{-}T_{f_0})]^{\nu}$ (where ν is a critical exponent) when the critical temperature T_{f_0} is approached from above, the expression for τ is given by [27]:

$$\tau = \tau_0^* \left(\frac{T - T_{f_0}}{T_{f_0}} \right)^{-z\nu}. \tag{2}$$

This relation for τ, when recast in terms of the measurement frequency f, yields:

$$\tau = \tau_0^* \left(\frac{T_f - T_{f_0}}{T_{f_0}} \right)^{-z\nu}, \tag{3}$$

where T_f corresponds to the peak in χ_1' at a given measuring frequency f and $\tau = 1/2\pi f$ [34].

The inset of Fig. 8 shows that Eq. (3) is followed over the entire frequency range with the values $\tau_0^* = 1.5(5) \times 10^{-13} s$, $z\nu = 6.0(1)$, and $T_{f_0} = 133.5(2)$ K. The value obtained for τ_0 falls within the range of other metallic spin-glass systems [35] and has been observed in magnetic cluster systems [27], whereas the $z\nu$ value compares well with those reported on different spin-glass systems ($5 < z\nu < 11$) [36], usually labeled as fragile regime. Overall, this value agrees with the one obtained through calculations by Ogielski and Morgenstern for three-dimensional spin glasses with short-range magnetic interactions ($z\nu = 7.9$), in contrast to the expected value in conventional phase transitions ($z\nu = 2$) [37]. Finally, the T_{f_0} value is consistent with the temperature of the $\chi_1'(T)$ peak.

In addition, in the critical slowing down description of the spin glass transition a dynamical scaling of the imaginary part of the susceptibility $\chi_1'(T)$ [38] is also expected:

$$\chi''(f,T) = \varepsilon^{\beta} F(f\varepsilon^{z\nu}), \tag{4}$$

where $\varepsilon = \frac{T-T_{f_0}}{T_{f_0}}$, β is the critical exponent corresponding to the order parameter and F is a universal function of its argument. Figure 8 shows the power-law scaling of $\chi_1''(T)$. The scaling has been made using the value of T_{f_0} and $z\nu$ obtained from the fits to Eq. (3). The result shows a reasonable scaling using $\beta = 0.4(1)$, with all the $\chi_1''(T)$ curves taken at different frequencies collapsing onto a universal curve. The β value is close to the exponent ($\beta = 0.5$) obtained through Monte Carlo simulations [27].

When subtle magnetic transitions are expected to appear in magnetic compounds one should take care of tiny details; this is the case when GP are present as those magnetic correlations in the PM are feeble and may be masked in the linear component of susceptibility (or even more in the irreversibility of the static magnetization). In consequence, we step forward and ascertain exactly the nature of the magnetic state within GP. To achieve this goal, we will show and discuss the nonlinear susceptibility which is valid to evaluate whether there is a coexistence of a FM with the (already evaluated above) cluster-glass transition. The nonlinear response of the magnetization M is described as a series of terms containing applied field powers, in an expansion:

$$M(h) = M_0 + \chi_1 h + \chi_2 h^2 + \chi_3 h^3 + ..., \tag{5}$$

where M_0 is the spontaneous magnetization, h the oscillating field $h = h_0 \sin \omega t$, χ_1 is the linear susceptibility, and χ_2, χ_3 are the nonlinear susceptibilities. Based on the temperature variations of the nonlinear susceptibilities χ_n ($n = 1$–5), an ideal spin glass can be unambiguously distinguished from a ferromagnet [39] and from a collection of nanometric particles [22]. In a ferromagnet, it is expected that all susceptibility terms contribute to the magnetization and the appearance of a peak at the Curie point. By contrast, for an ideal or canonical spin glass, the even harmonics in the magnetic response, i.e., the nonlinear susceptibilities χ_2, χ_4 are zero [39] at temperatures $T < T_f$ and $T > T_f$ because they are proportional to the spontaneous magnetization M_0 or its powers, and $M_0 = 0$ in both spin glass and paramagnetic phases.

The real component of the nonlinear susceptibility χ_2' as a function of temperature for selected frequencies is shown in Fig. 9. For the sake of clarity, data in the proximity of T_C (80–125 K) and T_{G^*} (125–165 K) are plotted separately [Figs. 9(a) and 9(b), respectively]. At a fixed frequency, a large peak is observed around the long-range FM transition ($T_C \sim 105$ K), whereas a double-peaked variation is observed in the 125–165 K temperature range. Both peaks (∼155 K and ∼140 K) are separated by a temperature span of ∼15 K. Those peaks are better defined in $\chi_3'(T)$ signal [see inset of Fig. 9(b)].

The observation of the χ_2' contribution in this temperature range indicates the presence of short-range ferromagnetic order and discards the *canonical* SG scenario. The short-range nature of the FM order is inferred from the broad anomaly (a transition width of about 20 K in the present case) that results when the spin-spin correlation length ξ does not

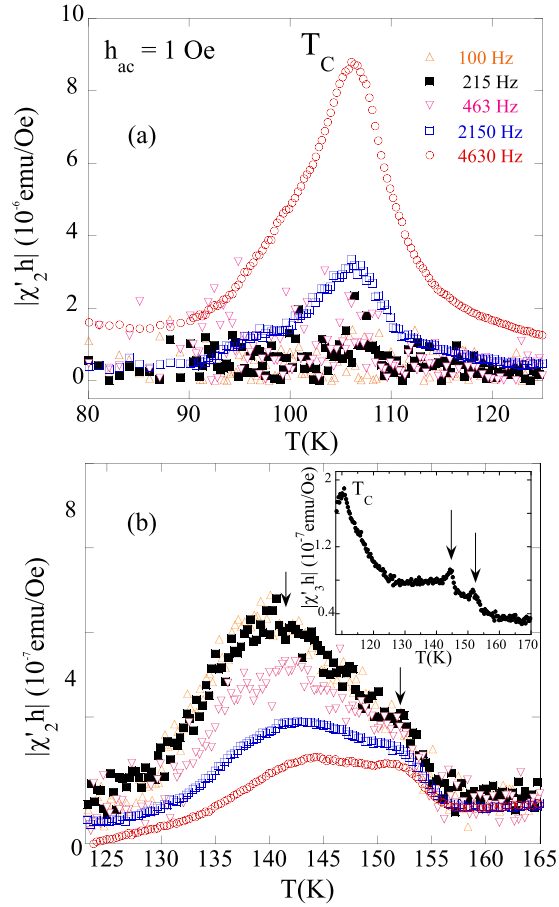

FIG. 9. Temperature variation of the nonlinear susceptibility χ_2' at various fixed frequencies when $H_{dc} = 0$ and $h_{ac} = 1$ Oe for (a) the temperature regime around T_C and (b) temperature regime around T_{G^*}. Nonlinear component χ_3' at a frequency of 10 kHz is displayed in the inset. T_{G^*} and the freezing transitions are marked by arrows.

diverge, but remains finite, at the critical temperature. The presence of such short-range FM correlations was evidenced by SANS on the parent compound $Tb_5Si_2Ge_2$ [13]. Those studies revealed the existence of FM-correlated regions of approximately 0.5 nm at $T_G \sim 200$ K, which grow in size on cooling reaching ∼5 nm at 165 K, where the size of the FM correlations abruptly rose beyond the experimental resolution of the instrument.

The presence of the characteristic experimental signatures of short-range FM order in $\chi_2'(T)$ for $x = 0.075$ indicates that at the time scale we are sampling (frequencies from 100 Hz to 10 000 Hz) the magnetic response is dominated by small ferromagnetic clusters developing in the GP. On the other hand, the two-peaked variation of $\chi_2'(T)$ and $\chi_3'(T)$ displayed in Fig. 9(b) is providing deeper information than that shown in Fig. 7 where only a broad maximum centered around 140 K in $\chi_1'(T)$ and $\chi_1''(T)$ was resolved. Here, the nonlinear components of the susceptibility enable the observation of a very fine magnetic process in this temperature range that it was not fully resolved by the linear susceptibility. The great sensitivity of these techniques has allowed us to detect subtle magnetic changes such as phase transitions or the presence of minuscule magnetic phases in heterogeneous nanomag-

netic systems such as the amorphous FeZr-based reentrant ferromagnets [40,41], reentrant spin-glass insulating systems ($La_{0.5}Sr_{0.5}CoO_3$ [25]), CMR oxides with cluster-glass spin structure $La_{0.7}Pb_{0.3}(Mn_{0.8}Fe_{0.2})O_3$ [41], the canted-spin system $Ce(Fe_{0.96}Al_{0.04})_2$ [42], and the hole-doped manganite $La_{0.7}Pb_{0.3}(Mn_{1-x}Fe_x)O_3$ [22], among others.

In the present case, the thorough analysis of ac susceptibility gives a detailed insight into the dynamics of freezing. It is crucial to point out that, unlike the examples mentioned above, the observed features in both the linear and nonlinear susceptibility occur above the well-defined PM-FM transition, i.e., in the paramagnetic regime, which is really striking. The peak in the nonlinear components $\chi_2'(T)$ and $\chi_3'(T)$ at ∼155 K [which is accompanied by a rise in $\chi_1'(T)$ and $\chi_1''(T)$, the irreversibility in the ZFC-FC magnetization and the stairlike fall in χ_{dc}^{-1} (T)] is necessarily due to a cooperative process where the enhancement of short-range FM correlations occurs, i.e., the recoupling of clusters previously formed at T_G. At lower temperature, the peak at ∼140 K in $\chi_2'(T)$ and $\chi_3'(T)$ [which is accompanied by the frequency dependent broad maxima in $\chi_1'(T)$ and $\chi_1''(T)$] is then associated with the freezing of clusters following the critical slow down process in our time scale of observation. The two-closely magnetic processes can explain the observed differences between T_{G^*} (∼155 K) as defined by χ_{dc}^{-1} (T) and the temperature at which the cluster-glass transition is observed by $\chi_1'(T)$ and $\chi_1''(T)$ (∼136 K).

The different magnetic nature of the ∼155 K and the ∼140 K peaks is supported by the frequency dependence of $\chi_2'(T)$ signal shown in Fig. 9(b). The dynamic response at ∼140 K gets modified drastically as ac frequency increases: The peak shifts to higher temperatures and its amplitude gradually diminishes with increasing frequency, giving rise to a broader relaxation. By contrast, the peak at ∼155 K is barely affected by ac frequency. Those differences suggest that a freezing process occurs at lower temperatures (∼140 K) whereas a cooperative process takes place at $T_{G^*} \sim 155$ K. This situation is clearly reminiscent to what it is established as a reentrant spin glass state. There, the magnetic moments become cooperatively coupled at the (phase) ordering transition and a long-range ferromagnetic order is clearly established. However, the presence of clusters (small domains) favor the appearance of a freezing at lower temperatures with a concomitant increase of the anisotropy and reduction of magnetization (and ac susceptibility) [27]. In the present case, this occurs at higher temperatures than the conventional ordering precisely in the GP. Certainly, this is unusual and the moment coupling in the GP above T_C act as a reentrant system. Some resemblance could be found with the metallic $4f$-based $CeNi_{1-x}Cu_x$ alloys as there was evidence of some clustering (also by SANS) above the magnetic ordering of the system [30,43]. Some (analytical) theoretical work has been dealing with the possible presence of magnetic clustering above a ferromagnetic state, in f-electron systems with an intense RKKY interaction. In that case, it was possible to propose useful magnetic phase diagrams [44,45]. Nevertheless, it is true that the inclusion of the Kondo interaction in the analytical Hamiltonian is not present in our $Tb_{5-x}La_xSi_2Ge_2$ system and hence the development of another more specific model would be welcome.

It is well known that lattice and magnetic structure of this family present a strong correlation. According to previous findings [13,14], T_G is marking the onset of the ferromagnetic clusters, i.e., the formation of an ensemble of nanometric regions of orthorhombic character $O(I)$ and strengthened FM interactions, due to structural defects of small deviations of the stoichiometric proportion of Si/Ge atoms. Several evidences suggest that a small substitution of Tb by a small amount of La modifies the magnetic response coming from the short-range FM clusters: (i) T_{G^*} is defined by a sharp drop in χ_{dc}^{-1} (T) in the $x = 0.075$ compound, whereas this step is much weaker in the parent compound $x = 0$, (ii) the anomaly in $\chi_1''(T)$ is much larger in magnitude in the $x = 0.075$ compound, and (iii) the transition manifests as a broad and large peak in $\chi_1'(T)$ for the $x = 0.075$, whereas it is not distinguished for the parent $x = 0$ compound. All these observations suggest overall an enhancement of the magnetic response of the FM clusters over the M-PM matrix in the diluted ($x = 0.075$) compound respect to the parent compound. Such an enhancement is connected to a larger typical FM cluster size and/or a larger number of FM clusters within the M-PM matrix in the diluted compound. The similar values obtained for μ_{eff} from dc susceptibility data in the different plateaus for both $x = 0.075$ and $x = 0$ compounds suggest the presence of clusters of similar sizes in both cases. Moreover, this hypothesis is further supported by the obtained values for the λ exponent in the GP which were very similar in both cases [$\lambda_G = 0.27(2)$ for $x = 0.075$ and $\lambda_G = 0.31(1)$ [13] for the parent compound $x = 0$].

Although the magnitudes of the nonlinear ac susceptibility peaks are tiny in this Griffiths-like compound, it is reasonable to provide a rough estimation of cluster size, as that can be immediately compared with the real value extracted from SANS in $Tb_5Si_2Ge_2$ [13]. For this, it is possible to take: $\zeta = r_{AV}\varepsilon^{-\nu}$ where ζ is the cluster spin-spin correlation length, r_{AV} is assumed to be the average nearest-neighbor distance between Tb^{3+} ions, ε is the reduced critical temperature, and ν is the dynamic exponent (see Bitla *et al.* [22] for details). Taking into account that $r_{AV} = 0.35(1)$ nm [9], $z \approx 10$ according to Bitla *et al.* [22] (which implies that $\nu \approx 0.6$ since $z\nu = 6$ in this system) and $\varepsilon \approx 0.1$ [22], then the above relation yields an average FM cluster size of $\zeta \approx 1.5(3)$ nm. This value is relatively close to the maximum value provided by SANS, where $\zeta \approx 5$ nm, although care should be taken as the characteristic frequencies involved in neutron ($\approx 10^{14}$ Hz) and ac susceptibility ($\approx 10^3$ Hz) are different. We therefore propose that the enhancement of the FM signal induced by La substitution should be explained in terms of a larger number of FM clusters within the PM matrix in the GP of the $x = 0.075$ compound.

The introduction of a nonmagnetic La atom substituting the magnetic Tb^{3+} tends to break the periodicity of the Tb atoms in the cell. This mechanism could give rise to more isolated regions of coupled magnetic moments with respect to the parent compound, which may serve as new nucleation seeds for additional regions with strengthened FM interactions (i.e., a large number of ferromagnetic clusters within the PM matrix). Furthermore, La is preferentially located at the R_2 site in the low La concentration compounds [18]. From the magnetic standpoint, this implies that La is not interfering with the FM interaction at the R_1-T_3-T_3-R_1 chain. However,

the La occupation of this site does interfere in the interactions between Tb atoms, since it favors the collinearity of the Tb magnetic moments (i.e., a decrease of AFM interactions between the Tb atoms). Thus, the random Tb/La distribution in the R_2 site introduces additional disorder in the magnetic AFM/FM interactions, leading to frustrated RKKY magnetic exchange interactions between Tb atoms, which is a key ingredient for the freezing process to appear.

IV. CONCLUSIONS

The static and dynamic susceptibility data presented in this paper provide clear evidence for the existence of magnetic relaxation resembling the standard cluster-glass state below a characteristic freezing temperature within the Griffiths phase in the magnetocaloric compound $Tb_{5-x}La_xSi_2Ge_2$ ($x = 0.075$). Nonlinear susceptibility response reveals that ferromagneticlike correlations are built up at 155 K, which become frozen at a lower temperature 140 K, thus in analogy with a reentrant spin glass behavior. Unlike the latter, this reentrant behavior does not occur below the long-range ordered state but in the paramagnetic regime within the GP. We suggest that the cluster-glass behavior in $x = 0.075$ originates from the structural disorder of the Tb and La atoms, which leads to frustrated RKKY magnetic exchange interactions between Tb atoms. Although no significant changes were observed in the magnetic (T_C, T_G) and structural properties for very low La doping levels ($x < 0.25$) [18], the experimental evidence obtained from our study shows that a small amount of La doping ($x = 0.075$) is able to affect the mechanism responsible for the formation of the short-range FM clusters within the M-PM phase in this system. In particular, we propose that La modifies the concentration of FM clusters within the M-PM matrix. Further investigations are required, such as SANS (as already performed in the parent compound) or muon-spin spectroscopy (μSR) in order to discern the different magnetic volume fractions within the paramagnetic phase. The existence of a magnetic (Griffiths) disorder state also opens up the possibility of a fine tuning of the magnetic exchange interactions via the annealing of the samples, which will result naturally in a modification of the clustering state.

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