

Griffiths-like phase of magnetocaloric $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ ($R=\text{Gd, Tb, Dy, and Ho}$)A. M. Pereira,^{1,*} L. Morellon,^{2,3} C. Magen,⁴ J. Ventura,¹ P. A. Algarabel,² M. R. Ibarra,^{2,3} J. B. Sousa,¹ and J. P. Araújo¹¹*IFIMUP and IN-Institute of Nanoscience and Nanotechnology, Departamento de Física da Faculdade de Ciências, Universidade do Porto, Rua do Campo Alegre, 687, 4169-007 Porto, Portugal*²*Instituto de Ciencia de Materiales de Aragón, Departamento de Física de la Materia Condensada, Universidad de Zaragoza and Consejo Superior de Investigaciones Científicas, 50009 Zaragoza, Spain*³*Instituto de Nanociencia de Aragón, Departamento de Física de la Materia Condensada, Universidad de Zaragoza, 50009 Zaragoza, Spain*⁴*Instituto de Nanociencia de Aragón-ARAID, Departamento de Física de la Materia Condensada, Universidad de Zaragoza, 50009 Zaragoza, Spain*

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We report on the discovery of Griffiths-like phase in several compounds of the $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ ($R=\text{Gd, Tb, Dy, and Ho}$) system, through the existence of an anomalous behavior on the reciprocal magnetic susceptibility (χ^{-1}). We found that this anomaly is restricted to the samples which present the $\text{Gd}_5\text{Si}_2\text{Ge}_2$ -type (monoclinic) or the Sm_5Ge_4 -type {orthorhombic-II [O(II)]} structural phases at room temperature. This peculiar effect originates from local disorder within the crystallographic structure, stabilized and enhanced by the competing intralayer and interlayer magnetic interactions. From this strong competition, a ratio of ~ 0.5 between Néel and the Griffiths-like temperatures is always found in the O(II) structure. In addition, a universal (x, T) phase diagram is proposed with an interpretation based on percolative processes.

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A strong correlation between structural and magnetic properties leads to the appearance of different exotic regimes in many magnetic systems (e.g., multiferroic, magnetic semiconductors, colossal magnetoresistive, and giant magnetocaloric compounds). This includes the *preformation* of ferromagnetically ordered nanometric clusters at some well-defined temperature T^* much above the true long-range ferromagnetic (FM) ordering at the Curie temperature T_C (Refs. 1–4) [i.e., still in the paramagnetic (PM) phase]. This regime between T^* and T_C ,^{5,6} was predicted by Griffiths in 1969 in diluted ferromagnets⁵ and has since attracted considerable interest.⁷ In its simplest form, the original problem considered the percolative nature of an Ising system⁸ having nearest neighbor (exchange) bonds characterized by a strength J occurring with a probability p ; otherwise, the bond strength is zero.^{6,7} For $p < p_c$ (percolation threshold), an infinite percolating *backbone* cannot be formed (or, equivalently, the correlation length does not diverge) and thus no cooperative FM transition occurs.⁶ Above p_c , the FM phase exists in a weakened form by the shortage of a percolation path; hence, thermal fluctuations will destroy the FM phase at a temperature T_C , which is lower than the critical temperature T_G ($=T^* > T_C$) of the *pure* FM phase (Griffiths temperature).⁸ The effect of disorder above T_C is to destabilize the pure system into small FM clusters. These small clusters give rise to characteristic features that allow the identification of the so-called Griffiths phase, namely, the deviation of the reciprocal susceptibility (χ^{-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.⁹

Experimental observations of the Griffiths phase were reported much later. A field-induced Griffiths phase was observed in FeCl_2 , manifesting itself as domainlike AFM correlations in the PM state.¹⁰ Further works claimed the presence of the Griffiths singularity in magnetic

semiconductors,^{11,12} intermetallics,^{13,14} oxides,^{2,4,9,12,15,16} and more recently, rare-earth (R) compounds.^{17–19} Within rare-earth systems, we highlight the $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ family. The observation of a Griffiths-like phase within this family of compounds was reported for $\text{Tb}_5\text{Si}_2\text{Ge}_2$.²⁰ The appearance of this regime has then pointed to arise from the strong interplay between structure and magnetism present in these materials. Their complex nanostratified crystalline structure with a unit cell constituted by two rigid slabs with five atomic layers each $[(\text{Si, Ge})/R/(\text{Si, R, Ge})/R/(\text{Si, Ge})]$, is the key parameter.²¹ At the interface between these two building block occurs the formation/rupture of $\text{Si}(\text{Ge})\text{-Si}(\text{Ge})$ dimers linked through covalent bonds.²² This process leads to the formation of three distinct structures: the orthorhombic-I [O(I)], the monoclinic (M) and the orthorhombic-II [O(II)]. The O(I) structure belongs to the $Pnma$ space group and is characterized by the fully formed bonds between the interslab $\text{Si}(\text{Ge})$ atoms. In the M phase (with a $P112_1/a$ space group) only half of $\text{Si}(\text{Ge})$ covalent bonds in the interslab region are formed, whereas the O(II) structure (also $Pnma$ symmetry group) presents all the $\text{Si}(\text{Ge})$ bonds at the interface of the two rigid slabs broken.²² Subsequent studies on the $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ compounds gave different insights on the properties of these clusters displaying short-range magnetic correlations (SRMCs). Studies on single-crystalline Gd_5Ge_4 showed the anisotropic character of such SRMC, being stronger along the b axis, i.e., along the $\text{Si}(\text{Ge})\text{-Si}(\text{Ge})$ interface bond direction.²³ For $\text{Tb}_5\text{Si}_{2.2}\text{Ge}_{1.8}$ single crystals, this anisotropy is stronger along the a axis.¹⁹ The presence of similar SRMCs were reported in polycrystalline $\text{Dy}_5\text{Si}_2\text{Ge}_2$ (Ref. 24) and also, very recently, on Ho_5SiGe_3 .²⁵ Inelastic neutron-scattering studies in Tb_5Ge_4 have been carried out to infer about the origin of such short-range correlations.^{26–28} Previous works on $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$ established that the Griffiths phase arises from the onset of spin clusters at the temperature of the *un-*

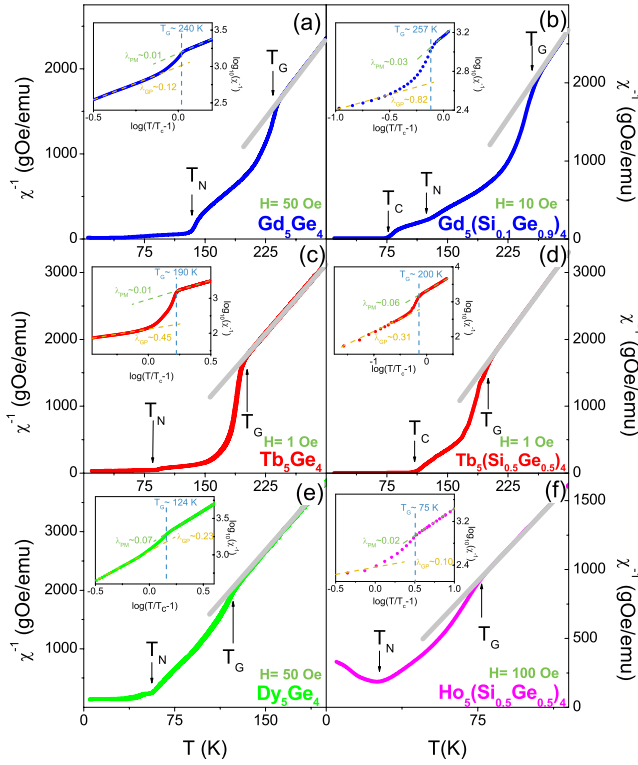


FIG. 1. (Color online) Temperature dependence of reciprocal susceptibility (χ^{-1}) of (a) Gd_5Ge_4 , (b) $\text{Gd}_5(\text{Si}_{0.1}\text{Ge}_{0.9})_4$, (c) Tb_5Ge_4 , (d) $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$, (e) Dy_5Ge_4 , and (f) $\text{Ho}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ compounds. Insets: linear fits of χ^{-1} vs $(\frac{T}{T_C}-1)$ in double logarithmic scale near T_C .

diluted magnetic phase (the critical temperature expected if the structure remains unchanged), identified as the O(I).²⁹ However, the parameter that controls this unexplored regime on the $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ systems remains to be explained. The main outcome of this Brief Report is the unraveling of the generic nature of these clusters in the compounds of the $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ family of magnetocaloric materials and the conclusion that there is a universal process ruling the magnetic properties.

Polycrystalline specimens of $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ systems were synthesized by arc-melting of stoichiometric mixtures of high-purity (99.9 wt %) R ($R=\text{Gd}, \text{Tb}, \text{Dy}$, and Ho) and (99.9999 wt %) Si and Ge. Weight losses during melting were negligible, and therefore the initial composition assumed unchanged. Further details on sample preparation can be found in Refs. 20, 30, and 31. The quality of the as-cast samples was checked by room-temperature x-ray diffraction and scanning electron microscopy. The crystallographic structure of all samples was refined using the x-ray diffraction analysis FULLPROF software,³² all alloys being single phase. Low-field dc magnetization measurements were carried out using a commercial (Quantum Design) superconducting quantum interference device magnetometer.

The low-field reciprocal susceptibility ($\chi^{-1} = \frac{H}{M}$) of selected $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ compounds, with $R=\text{Gd}, \text{Tb}, \text{Dy}$, and Ho , is shown in Fig. 1. The studied systems always exhibit an O(II) or M crystallographic phases at room temperature. From Fig. 1 one observes that, on cooling, χ^{-1} exhibits a

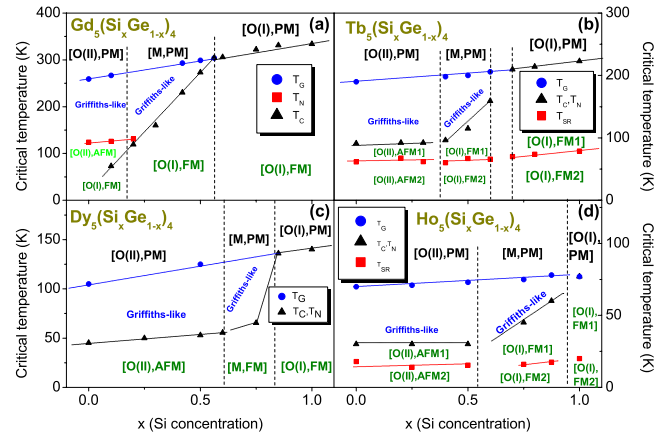


FIG. 2. (Color online) (x, T) phase diagram of the systems: (a) $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$, (b) $\text{Tb}_5(\text{Si}_x\text{Ge}_{1-x})_4$, (c) $\text{Dy}_5(\text{Si}_x\text{Ge}_{1-x})_4$ including data from Ref. 24 and (d) $\text{Ho}_5(\text{Si}_x\text{Ge}_{1-x})_4$ (notice that the temperature region of the formation of SRMC in Ref. 25 match well with our phase diagram).

linear thermal dependence for all samples (as expected from the Curie-Weiss law) that however disappears at a temperature T_G above the corresponding ordering temperature (Curie T_C or Néel T_N temperatures). A “stairlike” behavior is then observed in the $T_{C,N} < T < T_G$ intermediate region. Furthermore, a magnetic susceptibility exponent ($\beta = 1 - \lambda$) [$\chi^{-1}(T) = (\frac{T}{T_C} - 1)^{-\lambda}$; $\lambda > 0$] is obtained in this temperature range (see insets of Fig. 1). This remarkable behavior is typical of finite-size FM clusters in a paramagnetic matrix and is the hallmark of Griffiths singularities. Furthermore, the disappearance of the magnetic contribution arising from the SRMC with the applied magnetic field was observed in all samples (not shown) as previously seen in similar compounds.^{19,20,23,24}

Using the experimentally determined $T_G(x)$ dependence, we can now reconstruct the (x, T) magnetic and crystallographic phase diagrams of the $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ system. Figure 2(a) display such diagram for the Gd compounds. It is noteworthy that T_G extrapolates linearly toward the Si-rich compound region [O(I) structural phase]. This is also a clear fingerprint of Griffiths-like singularities, considering that the O(I) phase corresponds to the nondiluted system in the original Griffiths consideration, as previously suggested by Ouyang *et al.*²³ A similar result is obtained for the case of $\text{Tb}_5(\text{Si}_x\text{Ge}_{1-x})_4$ when one includes the determined $T_G(x)$ behavior [Fig. 2(b)]. To reconstruct the Dy phase diagram, including this regime [Fig. 2(c)], our results were merged with those of Nirmala *et al.*,²⁴ who also suggested the presence of SRMC with a $T_G \sim 123$ K for $x=0.5$. In the case of $\text{Ho}_5(\text{Si}_x\text{Ge}_{1-x})_4$, the magnetic properties were recently reported in Ref. 30, it was also found the existence of SRMC in the PM phase confirmed by the recent work performed by Singh *et al.*²⁵ The extended (x, T) phase diagram is depicted in Fig. 2(d).

From the four presented (x, T) phase diagrams (Gd, Tb, Dy, and Ho), three main conclusions can be drawn: the first is that Griffiths-like behavior always appear in the M and O(II) phases, for compositions below a characteristic concen-

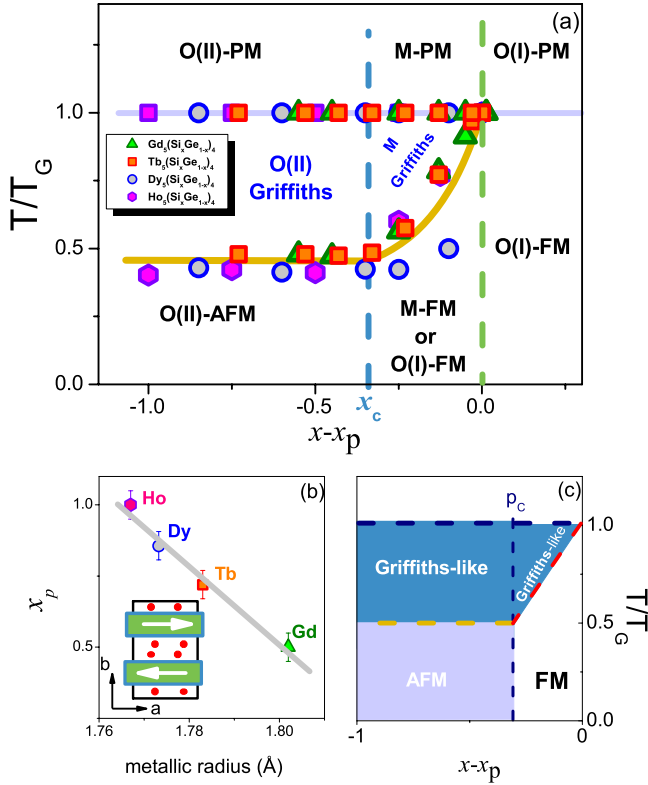


FIG. 3. (Color online) (a) Universal (x, T) phase diagram of the $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ compounds with $R=\text{Gd}$ (triangles), Tb (squares), Dy (circles), and Ho (hexagons). (b) x_p vs metallic radius of the rare earth. Inset: scheme of the magnetic structure of the O(II) crystallographic phase (c) Adapted T vs $x-x_p$ phase diagram of a dilute FM system for the $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ system.

tration (that we will here designate x_p), where T_C starts deviating from the $\frac{dT_C}{dx}$ slope of the stable O(I) phase. The second is that an approximately constant value of the $\tau = T_{C,N}/T_G$ ratio (~ 0.5) emerges in all systems when the O(II) structure is stabilized. The last outcome that can be inferred is that in the case of Gd , Tb , Dy and Ho , the O(II) structure is stabilized for $|x-x_p| < 0.33$.

Based on the aforementioned observations, we plot τ vs $(x-x_p)$ and verify a striking and intriguing collapse of all phase diagrams [Fig. 3(a)]. This finding allows us to suggest that the substitution of Si by Ge leads to the same effect in all systems, showing that the R element only shifts the value of x_p at which the SRMC starts to be detected. We propose this to be related with percolation effects, in full correlation with the nature of Griffiths phases,⁶ where the x_p concentration corresponds to $p=1$ in a regular ferromagnet. In $R_5(\text{Si}_x\text{Ge}_{1-x})_4$, two types of exchange interactions are present: one arising from the intraslab region J_{intra} , and the other from the interslab magnetic interactions J_{inter} . The latter, J_{inter} , largely depends on the distance between the rigid slabs, and has a FM character in the case of M and O(I) structures (where the slabs are partially or totally bonded), or AFM for the O(II) structure (when the slabs are unbonded). It is known that the substitution of Si by Ge in the M phase increases the interslab distance for the Gd system (e.g., Gd1-Gd2 distance)²² and leads to the rupture of the interslab

$T'-T'$ bonds, shortening the magnetic exchange pathways between the slabs since these covalent bonds present a magnetic polarization.^{21,33,34} These bond ruptures are thus the key feature for the appearance of the Griffiths-like phases in the described systems.

By taking into consideration the above results, we can understand the universal phase diagram of Fig. 3(a) and the nature of the Griffiths-like phases here observed by relating them with a percolation mechanism. When Si is being substituted by Ge in the interslab region, the break of the covalent bond is favored leading to a decrease in T_C . However, this process starts only at $x=x_p$, indicating that this is the critical concentration at which the Ge atoms start to occupy with higher probability the interslab sites. The observed linear dependence of the critical concentration on rare-earth ionic radius in Fig. 3(b) indicates that, with decreasing ionic radius, the empty space within the rigid layer decreases, thus decreasing the probability of the Ge atoms to be incorporated within the rigid slab. This picture fits perfectly with our results. In the case of $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$, Ge first occupies the intraslab sites and only afterward (at $x < x_p = 0.5$) the interslab positions. In contrast, for Ho compounds, Ge starts by first occupying the interslab sites and only afterward the rigid slabs ($x_p \sim 1$). A value of $x_c = |x-x_p| \sim 0.3$ is found for all phase diagrams [for which all $\text{Si}(\text{Ge})\text{-Si}(\text{Ge})$ bonds are broken; Fig. 3] suggesting that a percolationlike mechanism is responsible for the bond rupture (note that the theoretical value for a cubic lattice site percolation is $p_c \sim 0.31$). Based on theoretical considerations obtained in a previous work,³⁵ one can plot the expected phase diagram for systems displaying ferromagnetic short-range correlations, also including the AFM ground state usually presented in Ge -rich $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ compounds [Fig. 3(c)].³⁵ Furthermore, note that Burgý *et al.* showed that, in systems with a strong competition between two magnetic interactions (in our case, $J_{\text{intra}} > 0$ and $J_{\text{inter}} < 0$), a clean-limit critical ratio of 0.5 emerges in two-dimensional (2D) systems,³⁶ a value remarkably similar to that obtained for the τ parameter. In fact, one can consider that these materials are constituted by identical 2D subnanometer-thick rigid slabs interconnected by partially covalent bonds in the interslab [see inset Fig. 3(b)].³⁷ Finally, note that the existence of the two competing interactions was shown to lead to the enhancement of the FM short-range correlations.

In conclusion, we discovered the presence of short-range magnetic correlations in the M and O(II) structures for the $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ systems with $R=\text{Gd}$, Tb , Dy , and Ho . These correlations are an essential ingredient for the existence of Griffiths-like singularities, indicating that, when the system is in the M or O(II) phases, it retains memory of the O(I) phase (pure state) signaled by an anomaly at T_G . We further found a universal scaling of the phase diagrams for the $R_5(\text{Si}_x\text{Ge}_{1-x})_4$ systems, characterized by the $T_N/T_G \sim 0.5$ by the τ ratio (at which all bonds are broken) and the $x-x_p$ quantity, where x_p is the composition at which the Griffiths-like phase sets in. Moreover, the M phase appears within a universal composition window of $0.3 > x-x_p > 0$ clearly suggesting a percolation mechanism behind the observed universality. In order to more deeply understand the Griffiths-like

behavior observed in all compounds presenting the O(I) and M structure, further investigations are required, such as small-angular neutron scattering (as already performed in the $\text{Tb}_5\text{Si}_2\text{Ge}_2$ compound) or specific heat. By correlating these results with those obtained from magnetization measurements it will be possible to compare the emerging picture with other classes of important magnetic materials such as colossal magnetoresistive manganites.

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