Scientific highlights

**Observation of a Griffiths-like phase in the magnetocaloric compound Tb₅Si₂Ge₂**

Within the so-called Griffiths model, a magnetic system in which a random distribution of the magnetic interactions is induced by disorder leads to a situation where different values of the exchange constant can be randomly assigned to the different sites of the lattice. In a certain temperature range above the ordering temperature $T_C$, the disordered system presents an intermediate regime, called the Griffiths phase, between the conventional paramagnetic phase and the ferromagnetic state characterised by a cluster-like state.

The rare-earth intermetallic system $\text{Tb}_5(\text{Si}_{x}\text{Ge}_{1-x})_2$ was chosen as a serious candidate to host a Griffiths-like phase. The physical properties of these systems are strongly determined by their intrinsically layered crystallographic structure and the strong interplay between the magnetic and structural degrees of freedom. The crystal structure is defined by the stacking of rigid two-dimensional layers (slabs) of Tb and Tb + Si/Ge atoms. The actual crystallographic phase and the nature of the magnetic interactions are controlled by the number of interlayer T-T covalent-like bonds connecting the slabs [1], as these allow two different magnetic interactions to play part: the intralayer interaction ruled by the conventional 4f-4f RKKY indirect exchange, and the interlayer interactions, strongly influenced by an additional Tb-T-T-Tb superexchange-like interaction via the existing T-T bonds. Microscopic experimental evidence has been reported supporting that the intralayer magnetic structure is essentially ferromagnetic (FM), whereas the interlayer coupling can be either FM or antiferromagnetic (AFM) [2]. In the case of $\text{Tb}_5\text{Si}_2\text{Ge}_2$, the high-temperature paramagnetic (PM) state crystallises in a monoclinic $P1\overline{1}2_1/a$ structure. On cooling, this compound experiences a second-order transition to an FM state at $T_C = 110$ K, decoupled from the structural transformation to an

![Graph](image.png)

Figure 1: Temperature dependence of the inverse susceptibility ($\chi^{-1}$) as a function of magnetic field, measured on heating. The inset shows the temperature and field dependence of $\chi^{-1}$ calculated from the Lorentzian fits of the SANS spectra.

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**Figure 1:** Temperature dependence of the inverse susceptibility ($\chi^{-1}$) as a function of magnetic field, measured on heating. The inset shows the temperature and field dependence of $\chi^{-1}$ calculated from the Lorentzian fits of the SANS spectra.
The existence of a Griffiths-like phase in TbSi₂Ge, in the form of an FM cluster system within a PM matrix, is firstly indicated by macroscopic magnetisation experiments. Figure 1 presents the inverse dc magnetic susceptibility ($\chi'$) as a function of temperature on heating in low magnetic fields. This picture clearly illustrates the anomalous behavior of $\chi'$ that represents the fingerprint of a Griffiths phase [3]. In the conventional PM regime ($T > 200$ K), the effective paramagnetic moment is $9.8(1) \mu_B/tb$, which perfectly agrees with the theoretical value. However, below 200 K, a dramatic stair-like fall of $\chi'$ is evident at very low fields (< 100 Oe). On further increasing the magnetic field, only one plateau is observed, and the magnetic susceptibility at $H = 500$ Oe becomes indistinguishable from the high-temperature values. It has been shown that the Griffiths phase is univocally characterised by a magnetic susceptibility exponent lower than unity, i.e. $\chi' \propto (T_c - T)^{-\lambda}$, where $0 < \lambda < 1$ [4].

We have fitted the logarithmic representation of $\chi'$ obtaining clearly different values for the exponent $\lambda$, depending on whether we refine it in the anomalous region of $\chi'$ ($\lambda_\text{an} = 0.31$) or in the conventional PM phase ($\lambda_\text{PM} = 0.064$). The SANS instrument D16 is adequate to accurately probe the existence of FM clusters and characterise their size and temperature and magnetic-field evolution. The temperature dependence on cooling of the SANS intensity as a function of magnetic field, and at $Q = 0.1 \AA^{-1}$, which is a typical intermediate value in the range of transferred momentum within the resolution of the instrument, is shown in figure 2. First, a remarkable increase of the SANS signal is observed from 200 to 175 K, this step being followed by a small plateau that extends down to ~150 K. This anomalous contribution in the temperature range 150-200 K could be related with the nucleation of FM clusters within the PM region of TbSi₂Ge, below $T_c = 150$ K, a huge increase of the signal is found associated with a strong rise of magnetic correlations in the vicinity of the Curie temperature of a second-order FM transition. A double peak is seen, at $T_c = 115$ K and $T_T = 105$ K, which is associated with the decoupled magnetic-crystallographic transformation. Upon application of a magnetic field, the decrease of the SANS intensity in the whole temperature range is considerable. The correlation length $\xi$ extracted from the Lorentzian fits of the SANS spectra as a function of temperature and in different magnetic fields (see inset in figure 1) is the final proof of the existence of the FM cluster distribution that characterises the Griffiths-like phase. In zero-field, sizeable correlation lengths (~5 Å) are found around 200 K, its size progressively increasing up to a maximum at around $T = 165$ K, where the cluster size rises beyond the experimental resolution of the instrument. SANS experiments demonstrate the existence of FM clusters within the monoclinic PM phase of TbSi₂Ge, which characterise a Griffiths-like phase in the temperature range $T_c < T < 200$ K. It is suggested [5] that the Griffiths-like phase originates from the intrinsic disorder within the crystallographic structure, and the competition of the intra- and interlayer magnetic interactions ruling the microscopic behaviour of the naturally-layered structure, both factors promoting the segregation of nanometric regions with stronger FM interactions.

**References**