Local character of the highest antiferromagnetic Ce-system \( \text{CeTi}_{1-x}\text{Sc}_x\text{Ge} \)

J.G. Sereni, P. Pedrazzini, M. Gómez Berisso, A. Chacoma, S. Encina

Low Temperature Division, CAB-CNEA and CONICET, 8400 San Carlos de Bariloche, Argentina

T. Gruner, N. Caroca-Canales, C. Geibel

Max-Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany

(Dated: November 3, 2014)

The highest antiferromagnetic (AFM) temperature in Ce based compounds has been reported for CeScGe with \( T_N = 47 \) K, but its local or itinerant nature was not deeply investigated yet. To shed more light into this unusually high ordering temperature we have investigated structural, magnetic, transport and thermal properties of \( \text{CeTi}_{1-x}\text{Sc}_x\text{Ge} \) alloys within the range of stability of the CeScSi-type structure: \( 0.25 \leq x \leq 1 \). Along this concentration range, this strongly anisotropic system presents a complex magnetic phase diagram with a continuous modification of its magnetic behavior, from ferromagnetism (FM) for \( 0.25 \leq x \leq 0.50 \) (with \( 7 \leq T_C \leq 16 \) K) to AFM for \( 0.60 \leq x \leq 1 \) (with \( 19 \leq T_N \leq 47 \) K). The onset of the AFM phase is associated to a metamagnetic transition with a critical field increasing from \( H_{cr} = 0 \) at \( x = 0.55 \) to \( \approx 6 \) Tesla at \( x = 1 \), coincident with an increasing contribution of the first excited crystal electric field doublet. At a critical point \( x_{cr} = 0.65 \) a second transition appears at \( T_L \leq T_N \). In contrast to observations in itinerant systems like CeRh\(_2\)Si\(_2\) or CeRh\(_2\)B\(_2\), no evidences for significant hybridization of the \( 4f \) electrons at large Sc contents were found. Therefore, the exceptionally large \( T_N \) of CeScGe can be attributed to an increasing RKKY inter-layer interaction as Sc content grows.

I. INTRODUCTION

Magnetic order in Cerium compounds can be found along four decades of temperature, from 1.6 mK in CMN [1] up to 115 K in CeRh\(_2\)B\(_2\) [2], though most transitions occur between a few degrees and \( T_{ord} \approx 12 \) K. Different magnetic behaviors characterize different temperature ranges of magnetic order. While magnetic interactions between localized Ce-4\( f \) moments are observed for \( T_{ord} < 12 \) K [3], in the cases where \( T_{ord} \) exceeds that temperature, two types of behaviors can be distinguished. The one with localized Ce-4\( f \) moments is related to the rare cases of Ce binary compounds formed in BCC structure. This is the crystalline structure with the highest symmetry that allows to have the quartet \( \Gamma_8 \) as ground state. However, its four fold degeneracy is reduced by undergoing different types of transitions mostly connected with structural modifications. Those compounds are: CeZn \( (T_N = 30 \) K); CeTl \( (T_N = 25.5 \) K); CeMg \( (T_C = 20 \) K); CeCd \( (T_C = 16.5 \) K) and CeAg \( (T_N = 15.6 \) K) [3].

The ternary Ce-based compounds belonging to the other group present evidences of itinerant character [5]. Among them, the outstanding case is the mentioned CeRh\(_2\)B\(_2\) that shows the highest ordering temperature with \( T_{ord} = 115 \) K [2]. The following highest ordering temperature has been reported for CeScGe with \( T_{ord} = 47 \) K [6], but the local or itinerant nature of its magnetic state is under discussion. Within this group, one of the most studied compounds is CeRh\(_2\)Si\(_2\), which shows a \( T_{ord} = 36 \) K [7, 8]. Also in this case evidences for local and itinerant magnetic character were equally claimed by different authors [9, 11].

Such ambiguity is not unusual in Ce compounds and it has been discussed since the early studies on pure Ce under pressure and identified as the local-itinerant dilemma of Ce-4\( f \) electrons [12], not being completely elucidated yet. The eventual itinerant character of the 4\( f \) orbitals was compared to the U-5\( f \) behavior [9], whose compounds frequently show high \( T_{ord} \) values with clear itinerant characteristics arising from the extended character of the 5\( f \) orbitals. In Ce-4\( f \) systems, however, itineracy is related to some degree of hybridization with the conduction band and the consequent weakening of the 4\( f \) effective magnetic moment \( \mu_{eff} \). The question arises whether there is an optimal combination of weak \( 4f \)-band hybridization that enhances the inter-site RKKY interaction by increasing the in-site 4\( f \)-band exchange without reducing \( \mu_{eff} \) significantly.

The most convenient systems to investigate this problem are those showing high \( T_{ord} \) accompanied by experimental evidences where itineracy or localization character may emerge. In the case of CeScGe, it was formerly reported as a ferromagnet (FM) [5] that usually implies local 4\( f \) character. However, band-structure calculations [13] suggested that FM and antiferromagnetic (AFM) states competite in energy based in an itinerant character. Later on, a more detailed study on CeScSi and CeScGe [14] recognized both compounds as AFM. Thus, CeScGe presents the best characteristics for the proposed in-
vestigation.

A further aspect has to be taken into account for a proper knowledge of the ordered phase at those temperatures, which is the role of the excited crystalline field (CEF) levels. This contribution can be properly evaluated by tracing $T_{\text{ord}}$ from relatively low values, where only a doublet with local-$4f$ character contributes to the magnetic ground state. These conditions are fulfilled by CeTi$_{1-x}$Sc$_x$Ge alloys, that cover an extended range of transition temperatures from $T_{\text{ord}} \approx 7$ K in CeTi$_{0.75}$Sc$_{0.25}$Ge to $T_{\text{ord}} \approx 47$ K at the stoichiometric limit CeScGe \cite{6}. The lower $T_{\text{ord}}$ value is determined by the limit of stability of the CeScSi-type structure (at $x = 0.25$) on the Ti rich side. Below $x \approx 0.15$ this alloy stabilizes in the CeFeSi-type structure.

II. EXPERIMENTAL DETAILS AND RESULTS

Polycrystalline samples of CeTi$_{1-x}$Sc$_x$Ge with $0.25 \leq x \leq 1$ were synthesized by arc melting under Ar atmosphere the nominal amounts of the constituents, purity above 99.99%. The samples were turned over and remelted several times to ensure homogeneity. Then, the samples were placed in a tungsten boat wrapped with zirconium foil and annealed at 1200 °C for one week. The quality of the samples was verified by means of X-ray powder-diffraction measurements using Cu-K$_{\alpha_1}$ radiation ($\lambda = 1.54056 \text{ Å}$) in a Stoe-Stadip-MP diffractometer. The pattern was indexed on the basis of the tetragonal CeScSi-type structure.

Specific heat was measured between 0.5 and 50 K using a standard heat pulse technique in a semi-adiabatic He$^3$ calorimeter. The magnetic contribution $C_m$ is obtained by subtracting the phonon contribution extracted from LaTi$_{0.5}$Sc$_{0.5}$Ge. Magnetization measurements were carried out using a Quantum Design MPMS magnetometer operating between 2 and 300 K, and as a function of field up to 50 kOe. Electrical resistivity was measured between 2 K and room temperature using a standard four probe technique with an LR700 resistive bridge.

A. Structural properties

The CeTi$_{1-x}$Sc$_x$Ge system forms in two related crystal structure: CeFeSi-type for low Sc content (up to $x = 0.15$) and CeScSi-type beyond $x = 0.23$ \cite{16}. In the latter, each second Ce-double layer is shifted by (1/2, 1/2) with a rearrangement of Sc and Si atoms, becoming a body centered tetragonal instead of primitive tetragonal of CeFeSi, see Fig. 1 with the consequent doubling of the ‘c’ lattice parameter.

The unit cell volume dependence on Sc concentration and the ‘c/a’ ratio of the tetragonal lattice parameters are shown in Fig. 2. The increase of the unit cell volume can be explained by the larger atomic volume of Sc with respect to Ti, whereas the reduction of the ‘c/a’ ratio is due to the increase of the ‘a’ parameter because ‘c’ remains practically unchanged. This indicates that an expansion in the basal plain of the tetragonal structure occurs whereas the inter-layer distances in the ‘c’ axis direction are slightly affected.

B. Magnetic susceptibility

In Fig. 3 the inverse of the high temperature magnetic susceptibility (1/$\chi$) is presented after subtracting a Pauli type paramagnetic contribution $\chi_P$. This temperature independent contribution, already reported for CeScGe \cite{17}, is observed to decrease from $\chi_P = 0.910^{-3}$ at $x = 0.4$ to $0.4710^{-3}$ emu/Oe mol at $x = 1$, as shown in the inset of Fig. 3. The 1/$\chi$ dependence with concentration (evaluated for
within the range where a maximum of $\chi(20 \leq T_{ord} \leq 50 K)$ indicates the AFM character of the transition for samples with $x \geq 0.60$. The samples with $x \leq 0.5$ are included in the inset of Fig. 4 using a more extended $M(T)$ scale because of a FM contribution. The typical $M(T)$ dependence of a FM is observed for $x \geq 0.25$ and 0.3 alloys, whereas those with $x \geq 0.4$ show an incipient AFM component even in a field cooling procedure. The thermal dependence on the paramagnetic phase is compared in the main figure with a general Curie-Weiss function $\chi(T) = 0.56/(T - 15.5)\text{emu/Oe mol}$ (notice the positive $\theta_p = 15.5 K$) to remark the stability of the paramagnetic phase along the full concentration range. On the contrary, the ordered phase shows a continuous variation of $\chi(T < T_N)$ including the presence of a weak maximum at $T = T_L$ between $0.65 \leq x \leq 0.80$.

Those different behaviors can be sorted into three ranges of Sc concentration: i) for $x \leq 0.6$ the $M(T)$ dependence indicate an increasing mixture of FM and AFM components in the GS, with the ordering temperature increasing from $T_{ord} = 7 K$ at $x = 0.25$ up to 16K at $x = 0.60$. ii) At $x = 0.65$ the $\chi(T)$ maximum displays two shoulders which can be distinguished after a detailed analysis of the $\chi(T)$ curvature, i.e. its second derivative $\partial^2\chi/\partial T^2$. This feature reveals that the $x = 0.65$ concentration is placed very close to a critical point. iii) Between $0.65 \leq x \leq 0.8$, a slight kink appears at $T = T_L(x)$, below $T_N$, as seen in Fig. 4. Within this range of concentration, $T_N$ increases from 19 K at $x = 0.65$ up to

$T > 50 K$ indicates a decrease of Ce effective magnetic moment from $\mu_{eff}(x) \approx 2.25 \mu_B$ at $x = 0.4$ to $\approx 2 \mu_B$ at $x = 0.7$. Beyond that concentration $\mu_{eff}(x)$ remains practically unchanged. Notably, the paramagnetic temperature $\theta_p(x)$ is positive along the full concentration range and even increases from $\theta_p \approx 8 K$ at $x = 0.4$, up to 19 K at $x = 0.8$. At that concentration it slightly decreases down to 17 K at $x = 1$ as shown in the inset of Fig. 3.

A detailed variation of $\chi(T)$ around $T_{ord}$ is shown in Fig. 4 in a double logarithmic representation.
35 K at $x = 0.8$, whereas $T_L(x)$ increases from 18 K up to 26 K. iv) Above $x = 0.9$, $T_N$ keeps growing from 38 K at $x = 0.9$ up to 47 K at $x = 1$, whereas $T_L(x)$ is hardly seen in $\chi(T)$ measurements.

C. Electrical resistivity

The thermal dependence of the electrical resistivity $\rho$, normalized at 300 K, are presented in Fig. 5 for samples between 0.30 $\leq x \leq$ 1.0. The continuous increase of the $\rho(T)$ slopes indicates the weakening of Kondo type scattering with growing Sc content. In the inset of that figure, a detail of the logarithmic dependence of samples on the Ti-rich side (0.23 $\leq x \leq$ 0.60) normalized at 100 K are collected to better analyze the role of the Kondo effect. A logarithmic increase of $\rho(T)$ approaching the magnetic transition from high temperature is observed at $x = 0.23$ as an indication of Kondo scattering contribution. However, the intensity of this electronic scattering rapidly decreases with Sc concentration till to vanish at $x = 0.60$. At that concentration, the weakening of the $\rho(T)$ anomaly approaching the stoichiometric limit can be attributed to the variation of the Fermi level within the energy gap associated to the AFM state. As Ti$^{4+}$ atoms are replaced by Sc$^{3+}$ ones, the chemical potential decreases and eventually approaches the lower energy side of the gap. This simple picture has to be considered within the complexity of the Fermi Surface in a real system with many conducting electrons.

D. Specific heat

Specific heat results show several peculiar features along the full concentration range in coincidence with the different regimes observed in magnetic measurements. The results obtained on the 0.25 $\leq x \leq$ 0.65 alloys are presented in Fig. 6 as $C_m/T$, where $C_m$ indicates the magnetic contribution to the specific heat after phonon subtraction. Within the experimental dispersion, the maxima of $C_m(x)/T$ coincide with those observed in $M(T)$. A common feature along this concentration range is the lack of a $C_m$ jump at $T = T_{ord}$. Instead, a broadened mean field transition is observed, followed by a tail in $C_m/T(T > T_{ord})$ that reveals a significant contribution of magnetic correlations as precursors of the magnetic transition. This tail becomes more extended in temperature with increasing $T_{ord}(x)$ up to $x = 0.50$. Samples with $x = 0.60$ and $x = 0.65$ recover the slope above $T_{ord}$ together...
with a moderate increase of the $C_m/T$ maximum. This change of tendency coincides with the modification of the magnetic structure around $x = 0.50$. In this concentration range, the maximum of $C_m/T$ decreases as $\propto 1/T_{ord}$. Although such a decrease is a consequence of plotting $C_m(T)/T$, it reveals that $C_m(T_{ord})$ slightly decreases with decreasing $T_{ord}$ and does not extrapolate to zero according to the law of corresponding states.

An increase of the low temperature curvature in $C_m/T(T)$ indicates a progressive opening of a gap of anisotropy ($\Gamma$) in the magnon spectrum with concentration. To evaluate that evolution, the low temperature $C_m/T$ dependence was analyzed using the function $C_m/T = \gamma_0 + B \ast T \ast \exp(-\Gamma/T)$ as shown in Fig. 7 for samples with $x = 0.65$ and 0.8. The values computed for those concentrations are $\Gamma = 7\text{ K}$ and $\Gamma = 15\text{ K}$ respectively. Concerning the $\gamma_0(x)$ dependence, it drops from $\gamma_0 = 0.22\text{ J mol}^{-1}\text{K}^{-2}$ at $x = 0.25$ down to $\approx 0.02\text{ J mol}^{-1}\text{K}^{-2}$ at $x = 0.60$.

The main characteristic of the alloys with $x > 0.65$ is the split of $C_m/T$ into two maxima, in agreement with the $\chi(T)$ results. While the lower transition (at $T = T_L$) shows a cusp-like anomaly between $x = 0.7$ and 0.9, the upper one ($T_N$) is associated to a jump in $C_m/T(T_N)$. The cusp at $T = T_L$ can be related to the vicinity of the critical point accounting that in these polycrystalline samples any eventual first order transition could be broadened in temperature because of the random direction distribution of crystals and some eventual concentration inhomogeneities.

For $x \geq 0.9$, a further change in $C_m/T(T)$ is observed as depicted in the lower part of Fig. 7. The broad shoulder of $C_m/T(T)$ in samples $0.9 \leq x \leq 1$ can be attributed to the increase of the GS degeneracy from $N_{eff} = 2$ to 4 as the contribution of the first exited CEF doublet gradually increases. Also both transitions change their shapes because, while the one at $T_L$ transforms into a step like anomaly, that at $T_N$ becomes sharper and grows significantly. Notice that the $\Delta C_m(T_N)$ jump in $x = 1$ is $\approx 16\text{ J mol}^{-1}\text{K}^{-1}$, in between the values predicted in a mean field approximation for a doublet ($\Delta C_m = 1.5\tilde{R} = 12.5\text{ J mol}^{-1}\text{K}^{-1}$) and a quartet ($\Delta C_m = 2.2\tilde{R} \approx 18\text{ J mol}^{-1}\text{K}^{-1}$). The small anomaly observed in $x = 0.9$ at $T \approx 7\text{K}$ can be attributed to an extrinsic contribution of a small amount of Ce-oxide.

### III. DISCUSSION

#### A. Magnetization

The $M(H)$ hysteresis loops measured at $T = 1.8\text{K}$ on the alloys with $x \leq 0.5$ reveal the FM character of the ordered phase (see Fig. 8) concomitant with the increasing value of $M(x)$ measured at $H = 50\text{kOe}$ presented in Fig. 8. The saturation magnetization ($M_{sat}$), extracted from Fig. 8, as $M(x,H) = M_{sat} \times (1 - a/H)$, increases from 1.04$\mu_B/f.u.$ for $x = 0.30$ up to 1.15$\mu_B/f.u.$ for $x = 0.50$. At $x = 0.6$ $M_{sat}$ decreases to $1\mu_B/f.u.$ for $x = 0.60$, though this value is extracted from a reduced fitting range. Similarly, the coercive field increases from 1.1 kOe for $x = 0.30$ up to 2.6 kOe for $x = 0.50$.

At $x = 0.60$, the spontaneous FM magnetization is replaced by a small susceptibility, indicating a transition to an AFM state. Coincidentally, a metamagnetic transition (MMT) appears with the critical field $H_{cr}$ increasing with Sc concentration up to our experimental limit of $H = 50\text{kOe}$ with an initial ratio of $\partial H_{cr}/\partial x = 2.2\text{kOe/Sc\%}$. $M(H)$ measure-
to the eventual hybridization (i.e. Kondo effect) acting on the first excited level, the standard Schottky anomaly ($C_{Sch}$) may not describe the $C_m(T)$ dependence properly. Since a proper fit including usual hybridization effects ($V_{cf}$) between conduction and 4$f$ states acting on each excited level requires complex calculation protocols [18], a simple approach to mimic a broadening of the levels was applied. For this procedure the first CEF exited doublet at $\Delta_I$ is described using single Dirac levels equally distributed in energy around the nominal value of a non hybridized level. This method requires the strength of the hybridization to be smaller than $\Delta_I$, i.e. $V_{cf} < \Delta_I/k_B$. To properly account for the total magnetic contribution to the entropy, the second excited doublet (at $\Delta_{II}$) was included, without accounting for eventual hybridization effects since $\Delta_{II}$ largely exceeds the temperature range of this analysis. The applied formula is:

$$C_{Sch}(T) = \sum_i A_i \times [(\Delta_i/T) / \cosh(\Delta_i/T)]^2$$ (1)

The GS contribution was included after fitting the tail of $C_m$ at $T > T_{ord}$ with an arbitrary $f(a/T)$ function, being the total contribution to the specific heat: $C_{tot} = f(a/T) + C_{Sch}(T)$. In the inset of Fig. 6 the result of this fit to the experimental data is shown, including the detail of the $C_{Sch}$ function. The computed parameters are $\Delta_I = 35$ K and $\Delta_{II} = 155$ K, with an effective broadening of the first CEF doublet evaluated as $\delta = 12$ K. These results were checked with the evaluation of the magnetic contribution to the entropy involved in this Schottky anomaly, which for two excited doublets reaches $R \ln 3$. This analysis cannot be applied for higher Sc concentrations because the increase of $T_{ord}$ progressively approaches $\Delta_I$. Although no significant changes are expected in the CEF levels splitting, the reduction of the electrical charges at the transition metal sites from Ti$^{4+}$ [Ar4s$^2$3d$^2$] to Sc$^{3+}$ [Ar4s$^2$3d$^1$] may affect the strength of the CEF with a consequent variation of $\Delta_I$.

C. Entropy

The analysis of the thermal evolution of the magnetic contribution to the entropy $S_m(T)$ is shown in Fig. 9 normalized by the value of a Kramer’s doublet level: $R \ln 2$. This parameter provides complementary information to better understand this complex system. The alloy $x = 0.25$ with the lowest ordering temperature ($T_C = 7$ K) reaches $S_m = R \ln 2$ at $T = 11$ K, slightly above $T_C$, suggesting that only the GS doublet contributes to the magnetic order. However, since in the paramagnetic phase $S_m(T)$

![Figure 8](image-url)
increases continuously (i.e. without showing any plateau around $R\ln 2$) one may infer that the low energy tail of the first excited CEF level starts to contribute to the entropy at quite low temperature. This fact confirms the CEF level spectrum extracted in the previous subsection from the fit of $C_m(T)$.

Although the contribution of the first excited doublet may be marginal in the alloys with low Sc content, it becomes significant for higher concentrations as $S_m(T_{ord})$ increases with $T_{ord}(x)$. This feature becomes evident for $x \geq 0.5$ alloys because $S_m(T_{ord})$ clearly exceeds $R\ln 2$. Notice that around this concentration the AFM sets on. For $x = 1$ it practically reaches the value corresponding to two doublets, i.e. $R\ln 2$. This fact confirms the CEF level spectrum extracted in the previous subsection from the fit of $C_m(T)$.

The two relevant features are the FM to AFM change between $0.50 \leq x \leq 0.60$ and the critical point CP at $x \approx 0.65$. There, an intermediate phase sets in between $T_N(x)$ and $T_L(x)$. The full concentration range different regions were identified: i) between $0.25 \leq x \leq 0.50$ a FM-GS, determined by $M$ vs $H$ hysteresis loops of Fig. 6a, shows an increasing $M_{sat}$ that reaches its maximum value at $x = 0.50$. ii) A continuous change form FM to AFM occurs at $0.50 < x < 0.60$, evidenced by the MMT with $H_{cr}$ arising from zero. iii) At the CP ($x \approx 0.65$) a weak transition emerges at $T_L(x)$ below $T_N$ according to $C_m(T)$ and $\chi(T)$ results. Notably, $T_N(x)$ rises continuously up to $T_N = 47 K$ (at $x = 1$) despite of the weakening of the Ce effective magnetic moment.

All along the concentration range there is no indication for increasing hybridization effects between 4f and conduction electrons. On the contrary, some
evidence for Kondo effect like the increase in ρ(T) towards low temperature and an enhancement of γ₀ are observed on the Ti-rich side despite the FM behavior. However, both parameters indicate the weakening of this effect with concentration. Therefore, the high Tₙ observed in CeScGe cannot be attributed to any hybridization mechanism rather to an intrinsically strong RKKY interaction.

A general description of the magnetic evolution of this system can be proposed by considering each Ce-double-layer as nearly 2D-FM unit that weakly interacts with neighboring layers. The positive and even growing θₚ(x) values support an increasing intra-layer Ce-Ce neighbors FM interaction. Once the amount of Sc-3d electrons becomes relevant (at x ≈ 0.50) the inter-layers RKKY interaction takes over favoring an AFM stacking of the double-layers within an anisotropic but 3D scenario. The thermal population of the first CEF excited level also contribute to the FM - AFM change because of the modification of the effective moments.

Despite of ≈ 12% difference between Ti and Sc Goldschmidt metallic radii (rₜi = 1.46Å and rₛₑ = 1.64Å respectively), the Ce-T spacing (dₙₑ₋ₜ) only increases by ≈ 2% between CeTiGe (dₙₑ₋ₜᵢ = 3.45Å [20]) and CeScGe (dₙₑ₋ₛₑ = 3.53Å [14]) evaluated within the same CeScSi structure. This variation is in agreement with the slight change (≈ 1%) observed on the ’c’ axis between x = 0.30 and x = 1. Within a rigid sphere picture, this dₙₑ₋ₛₑ = 3.53Å and the sum of Sc and Ce metallic radii (rₛₑ = 1.64Å and rₙₑ = 1.86Å) become comparable. Therefore, a significant increase of the electronic overlap between Sc-3d and Ce-5d electrons can be expected with the consequent strengthening of the Ce-inter-layers RKKY interaction.

In this context one should notice that isotopic rare earth (R) compounds of the RTGe family also present very high ordering temperatures, like GdScGe (CeScSi type structure) that orders at Tₙ = 350K [21], more than a factor 7 higher than in CeScGe. Similar values are observed for GdTlGe (of CeFeSi type structure) with Tₙ = 412K [22]. These behaviors are in clear contrast with itinerant characters of the Ce-4f orbital.

IV. CONCLUSIONS

Apart from the significantly high ordering temperature of CeScGe at Tₙ = 47K, the main magnetic characteristics shown by this system are the continuous change from FM to AFM-I order and the presence of a CP point (at x = 0.65) associated to an intermediate AFM-II phase. These modifications of the magnetic ground state occur without affecting the local character of the Ce-4f orbital.

The layer type of the crystalline structure seems to play a basic role because it favors the formation of FM sheets involving two neighboring Ce planes. At low Sc content (0.25 ≤ x ≤ 0.50) the FM intralane RKKY interaction, resembling 2D-type order, dominates the scenario. The continuous increase of the positive θₚ temperature indicates that this Ce-Ce interaction even enhances with concentration.

After reaching the maximum of Mₘₙ at x = 0.50, the system develops a MMT transition from Hₙ₉ = 0. Such a continuous transition from a FM state to an AFM reveals the similar energy of these competing phases, that can be described as a smooth modification from a FM-stacking to an AFM-stacking of FM layers. This change in the inter-layers RKKY interaction can be driven by the variation of the electronic configuration of the intermediaries atoms Ti⁴⁺ and Sc⁴⁺.

The increasing inter-layers interaction strengthens the 3D character of the AFM phase evidenced by the Cₘ(T) jump at the magnetic transition. However, the intermediate phase AFM-II suggests that the magnetic wave vector in the ’c’ direction does not reach full commensuration at Tₙ. Nevertheless, the discontinuity in the order parameter reflected in the Tₙ transition decreases with growing the Sc content. The Sₘₙ(T,x) evolution confirms the first excited CEF level increasing contribution as Tₙ becomes comparable to Δ₁. Therefore, the record high ordering temperature of CeScGe has to be attributed to the convergence of different factors like: the Ce double-layer structure of FM character, the increasing RKKY interaction with Sc-3d¹ concentration and the vicinity of the first CEF excited level. All these conditions are related with the local characters of the Ce-4f orbitals.

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