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Charge-Doping driven Evolution of Magnetism and non-Fermi-Liquid Behavior in the Filled Skutterudite $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$

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The filled-skutterudite compound $\text{CePt}_4\text{Ge}_{12}$ is situated close to the border between intermediate-valence of Ce and heavy-fermion behavior. Substitution of Ge by Sb drives the system into a strongly correlated and ultimately upon further increasing the Sb concentration into an antiferromagnetically ordered state. Our experiments evidence a delicate interplay of emerging Kondo physics and the formation of a local $4f$ moment. An extended non-Fermi-liquid region, which can be understood in the framework of a Kondo-disorder model, is observed. Band-structure calculations support the conclusion that the physical properties are governed by the interplay of electron supply via Sb substitution and the concomitant volume effects.

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The interplay of localized and itinerant degrees of freedom is at the heart of strongly correlated systems and often results in the emergence of novel and unconventional electronic phases, like unconventional superconductivity or quantum-critical behavior. Addressing this strong-coupling problem remains a challenge, both experimentally and theoretically. The identification of new classes of intermetallic compounds that allow to study this emergence in a well-characterized setting is highly desirable.

The large family of filled-skutterudite compounds shows a wealth of topical physical phenomena [1–4]. The phases with general formula MT_4X_{12} are built up by a rigid covalently bonded transition-metal (T) pnictogen (X) framework with mostly ionically bonded filler atoms (M). The possible groundstates comprise various forms of magnetic ordering *i.e.* itinerant ferromagnetism and local anti-ferromagnetism, heavy-fermion behavior, superconductivity, half metallicity, and non-Fermi-liquid (NFL) behavior. This family of materials also has interesting properties for thermoelectric applications.

Recently, new filled skutterudites MPt_4Ge_{12} have been reported with M being Sr or Ba [5, 6], rare-earth metals La, Ce, Pr, Nd, Sm, Eu [6–8] as well as Th or U [9, 10]. Several MPt_4Ge_{12} compounds ($M = \text{Sr, Ba, La, Pr, Th}$) become superconductors with T_c up to 8.3 K [6]. For $M = \text{Nd, Eu}$ a well-localized nature of $4f$ electrons is observed [11] while $\text{SmPt}_4\text{Ge}_{12}$ features a heavy-fermion state at low temperatures [8]. Theoretical and spectroscopic studies have shown that states at the Fermi level (E_F) can be attributed mainly to Ge $4p$ electrons while Pt $5d$ states are lying rather deep and only partially form covalent bonds with Ge $4p$ [12, 13]. The electropositive filler elements M act as electron donors transferring charge to the $[\text{Pt}_4\text{Ge}_{12}]$ polyanion [13]. In these systems, in the absence of strong correlations, simple band-structure concepts can be used and have been successfully applied *e.g.* in the optimization of the super-

conducting T_c of $\text{BaPt}_4\text{Ge}_{12}$ by partial substitution of Pt by Au [14].

The role of electron-electron interaction in the Ce-based skutterudites has been of recent interest [3]. The question of how strong the electronic correlations are in these compounds and how to influence them by charge carrier doping and/or changing the unit cell dimensions through chemical substitution are as important as they are delicate. In this respect the filled Skutterudite compound $\text{CePt}_4\text{Ge}_{12}$ is particularly attractive, since previous studies have placed this skutterudite at the border between intermediate-valence and Kondo-lattice behavior [7, 15]. The low-energy properties of this system are described by an Anderson lattice model which, in the absence of charge fluctuations, reduces to the Kondo lattice model [16]. The nature of the quantum phase transition at the border of antiferromagnetism in the Kondo lattice is much debated [17, 18]. Only few examples are known where NFL effects have been observed in the intermediate-valence regime due to antiferromagnetic (AF) quantum-critical fluctuations, *e.g.*, in $\beta\text{-YbAlB}_4$ [19] or $\text{CeIn}_{3-x}\text{Sn}_x$ [20, 21].

In this Letter, we show that by a suitable atomic substitution we can tune $\text{CePt}_4\text{Ge}_{12}$ from a nearly intermediate-valence paramagnet through a NFL phase into an antiferromagnet of localized Ce- $4f$ moments. This phase sequence driven by the chemical substitution is discussed in the context of other NFL systems. Substitution of Ge by a larger isovalent element drives the system toward more localization, thus leading to a reduction of charge fluctuations. The obvious choice for this purpose is to replace or partially substitute Ge by Sn in $\text{CePt}_4\text{Ge}_{12-x}\text{Sn}_x$. This failed, since only small amounts ($x < 0.3$) of Sn could be substituted [22]. As alternatives, Sb or As, which are significantly larger than Ge and provide one extra electron, were explored [23, 24]. Contrary to Sn substitution, our studies demonstrate a large range of stability of $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$ of x up to 3.0

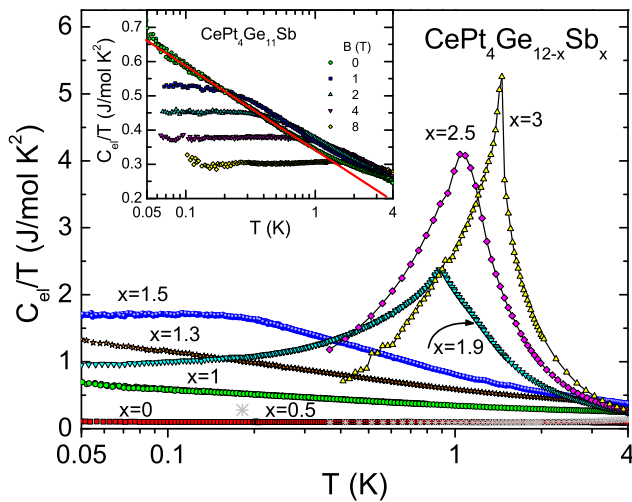


FIG. 1: (Color online) Electronic contribution to the specific heat, $C_{el}(T)/T$, of $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$ for different x . The low- T nuclear Schottky contribution to $C_p(T)$ has been subtracted. Inset: $C_{el}(T)/T$ of $\text{CePt}_4\text{Ge}_{11}\text{Sb}$ in different magnetic fields. At $B = 0$, $C_{el}(T)/T$ exhibits a logarithmic increase toward lowest temperatures (indicated by the red line), while $C_{el}(T)/T$ becomes constant in external magnetic fields.

in the filled-skutterudite structure.

Polycrystalline samples of $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$ were prepared by arc-melting stoichiometric amounts of the constituent elements. For homogenization, the samples were re-melted several times with a negligible mass loss. All samples were annealed at 820°C for 10 days and characterized by powder X-ray diffraction (XRD). The compositions of the observed phases were confirmed by energy dispersive X-ray spectroscopy (EDXS). Heat capacity and electrical resistivity experiments have been carried out utilizing a commercial measurement system (PPMS, Quantum Design) and a dilution refrigerator (Oxford Instruments). The magnetic susceptibility was determined in a SQUID magnetometer (MPMS, Quantum Design) equipped with a ^3He -option (iQuantum). The electronic structure of $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$ was studied using the full-potential local-orbital (FPLO) minimum basis code (version 9.01-35-x86) [25] within the local density approximation (LDA). In the scalar-relativistic calculations, the exchange-correlation potential of Perdew and Wang was employed [26].

In $\text{CePt}_4\text{Ge}_{12}$, Sb substitution on the Ge site expands the unit-cell volume. A linear dependence of the lattice parameter $a(x)$ in the $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$ series is observed (see below). In addition to the expansion of the unit cell, Sb substitution changes the chemical potential through electron doping.

The low-temperature properties of $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$ change strongly with Sb concentration. $\text{CePt}_4\text{Ge}_{12}$ has been shown to be close to intermediate-valence behavior [7]. Below $T_{\text{LFL}} \approx 8.4\text{K}$ $\text{CePt}_4\text{Ge}_{12}$ exhibits a Landau

Fermi-liquid (LFL) groundstate [7]. In slightly Sb-doped samples ($x = 0.5$) the magnetic and thermodynamic properties do not change qualitatively. The specific heat still indicates LFL behavior for $T < T_{\text{LFL}} \approx 7.9\text{K}$. On the other side, for high Sb concentration, ($x \geq 1.9$), we find AF order at low temperatures. In $\text{CePt}_4\text{Ge}_{10.1}\text{Sb}_{1.9}$, a sharp anomaly in the specific heat at $T_N = 0.89\text{K}$ indicates the magnetic ordering (see Fig. 1). Upon increasing the Sb concentration $T_N(x)$ shifts to higher temperatures. In $\text{CePt}_4\text{Ge}_9\text{Sb}_3$, with the highest Sb concentration of our study, $T_N = 1.46\text{K}$ is attained. The AF nature of the ordered phase is confirmed by magnetic susceptibility data, $M(T)/H$ (Supplemental Material Fig. S1), and is further supported by the magnetic-field dependence of T_N , which is suppressed to lower temperatures upon application of a magnetic field (Supplemental Material Fig. S2).

Magnetization measurements on the AF ordered samples ($1.9 \leq x \leq 3$) reveal that the magnetic moment does not saturate in a magnetic field up to 7 T. At 0.5 K only between $0.3\mu_B/\text{Ce}$ and $0.45\mu_B/\text{Ce}$ is reached, which is below the value expected for a Ce^{3+} doublet groundstate (Supplemental Material inset of Fig. S1). Even though the moment does not saturate in our experimentally accessible magnetic field range, the obtained values suggest a saturation moment which is not compatible with the small values typically observed in itinerant Ce systems. Also the magnetic entropy at T_N is reduced compared with $R \ln 2$ expected for a doublet groundstate [7]. From the magnetic specific heat, we obtain 23%, 30%, and 48% of $R \ln 2$ at T_N for $x = 1.9, 2.5$, and 3, respectively. The increasing magnetic entropy recovered at T_N indicates that the $4f$ moments are becoming more and more localized in character on increasing Sb concentration.

To get further insight in the electronic groundstate properties of the $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$ series we now turn to the electrical resistivity results presented in Fig. 2. The temperature dependence of ρ changes also drastically with Sb concentration. While in $\text{CePt}_4\text{Ge}_{12}$ ($\rho(T) - \rho_0$) $\propto T^2$ indicates LFL behavior below 10 K consistent with the specific heat [7], in $\text{CePt}_4\text{Ge}_{11.5}\text{Sb}_{0.5}$ a tiny increase of $\rho(T)$ toward low temperatures, becomes apparent below 3 K. This increase becomes more pronounced at $x = 1$, where $\rho(T)$ is best described by a logarithmic temperature dependence between 3.6 K and 0.75 K with a tendency to saturation at lowest temperatures. This contribution to the resistivity becomes more important with increasing x . Furthermore, a clear maximum in $\rho(T)$ develops for $x \geq 1.5$ (see Fig. 2). Upon increasing x , the position of the maximum [$T_{\text{max}}(x)$] shifts toward higher temperatures. In the magnetically ordered samples, T_N is marked by a small kink in $\rho(T)$ just below T_{max} . A summary of the experimental results is shown in Fig. 3c. The lines in this figure mark the general trend of T_N and T_{max} in x and are merely guides to the eye.

We note that both $T_{\text{max}}(x)$, as well as $T_N(x)$ seem to

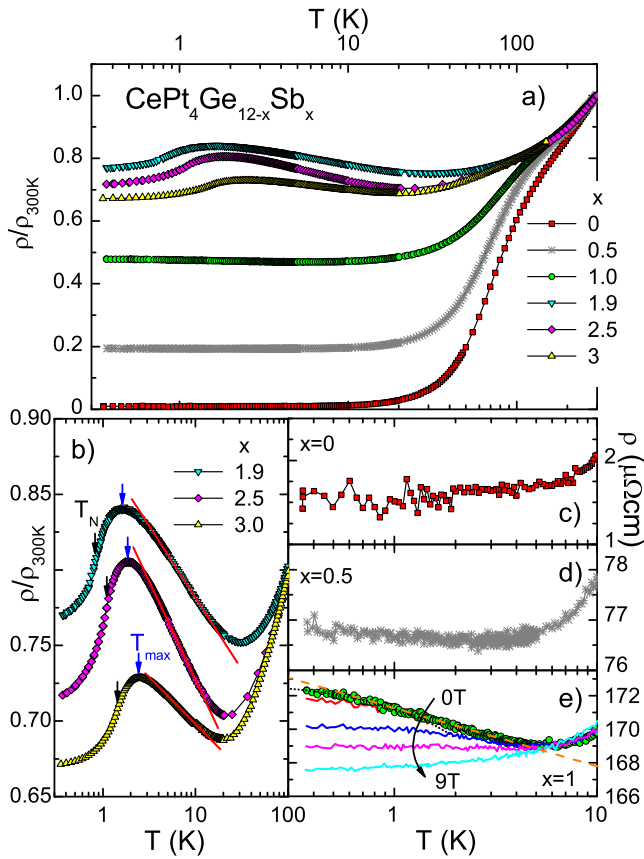


FIG. 2: (Color online) a) Normalized resistivity $\rho(T)/\rho_{300\text{K}}$ of $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$ for different x on a logarithmic temperature scale. b) Magnification for $x = 1.9, 2.5,$ and 3 . Straight lines indicate a logarithmic dependence of $\rho(T)/\rho_{300\text{K}}$ on T , reminiscent of incoherent Kondo scattering. The maximum and the kink in $\rho(T)/\rho_{300\text{K}}$ at T_{max} and at the Néel transition, T_N , respectively, are marked by arrows. c)–e) Low-temperature resistivity $\rho(T)$ for $x = 0, 0.5,$ and 1 . Except for $x = 0$ $\rho(T)$ increases toward low T . In e) the dashed line marks a logarithmic and the dotted line a linear temperature dependence of $\rho(T)$. In addition to 0 T (circles) data in magnetic fields of $1\text{ T}, 3\text{ T}, 5\text{ T},$ and 9 T (solid lines), are shown.

extrapolate to zero temperature in the region around $x = 1$. As a result, the phase diagram of $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$ resembles at first sight the generic phase diagram of a quantum-critical point (QCP) scenario separating a non-magnetic LFL state [7, 15] on one side from an antiferromagnetically ordered groundstate on the other side of the QCP. For $x = 1$ and 1.3 we find a logarithmic temperature dependence of C_{el}/T below 1 K . The data are shown in Fig. 1. A logarithmic divergence of C_{el}/T with temperature is also in line with general expectations for materials in close proximity to such AF QCPs. LFL behavior [$C_{\text{el}}(T)/T = \text{const.}$] is in both samples recovered in magnetic fields and extends to higher temperatures upon increasing field, at 8 T to well above 1 K . The inset of Fig. 1 shows the data for $x = 1$. Yet, the phase diagram in

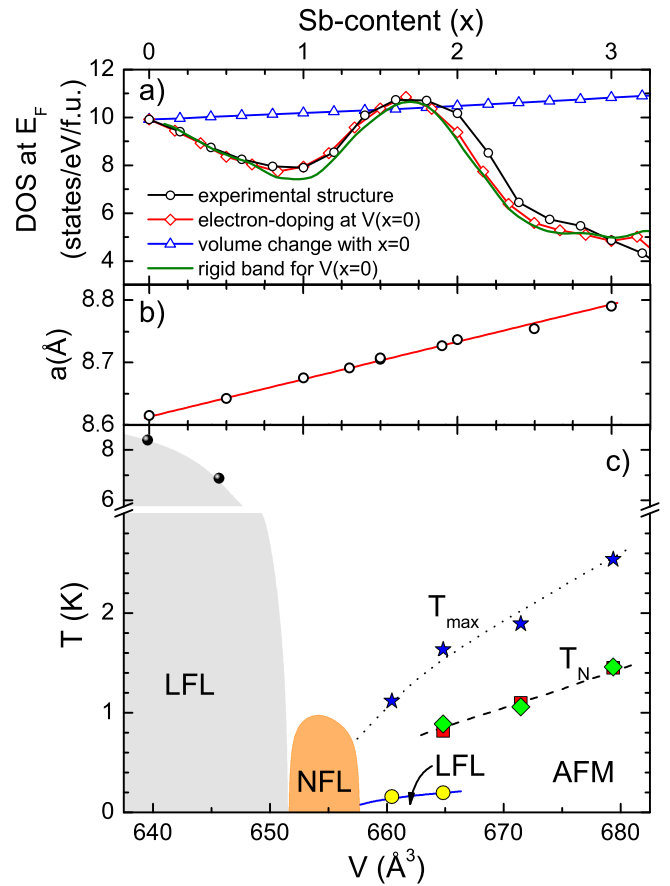


FIG. 3: (Color online) a) Density of states (DOS) at the Fermi energy (E_F) as function of the Sb concentration x calculated by different techniques/with different assumptions as indicated. b) Experimental lattice parameter $a(x)$ of $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$. (c) Temperature - unit-cell volume ($T - V$) phase diagram of $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$. The corresponding Sb concentration is indicated on the top axis. Samples $x = 0$ and 0.5 exhibit LFL behavior in an extended temperature range. AF ordering temperature T_N determined from specific heat (\blacklozenge) and from resistivity data (\blacksquare). Temperature of the maximum in the resistivity $\rho(T)$, T_{max} (\star). The samples with $x = 1$ and 1.3 show NFL behavior at low T . $\text{CePt}_4\text{Ge}_{10.5}\text{Sb}_{1.5}$ and $\text{CePt}_4\text{Ge}_{10.1}\text{Sb}_{1.9}$ display both LFL behavior below T_{LFL} (\bullet). The latter compound orders AF while $\text{CePt}_4\text{Ge}_{10.5}\text{Sb}_{1.5}$ does not show any magnetic order. The regions where LFL and NFL behavior is observed are indicated.

Fig. 3c, reveals some features that are incompatible with the standard AF QCP scenario: Even though the $T_N(x)$ line extrapolates to zero at $x \approx 1$, the NFL region is separated from the magnetically ordered phase by a non-magnetic LFL. At $x = 1.5$, $\gamma = C_{\text{el}}(T)/T|_{T \rightarrow 0} = 1.74\text{ J mol}^{-1}\text{ K}^{-2}$ becomes constant below 0.16 K , indicating a strongly correlated LFL groundstate, notably different from the one on the low Sb concentration side. The $C_{\text{el}}(T)/T = \text{const.}$ region extends to higher temperatures upon increasing magnetic field. Furthermore, we find no indication for a magnetic phase transition in any probe at

this Sb concentration. This extended NFL region and the separation of the magnetic phase from the NFL behavior cannot be explained within the generic QCP scenario.

Instead, the system evolves from a metal with moderate electron correlations which is located at the border to intermediate valence and a correspondingly high Kondo temperature into a local-moment magnet in an unconventional fashion. A full understanding of the $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$ phase diagram requires to disentangle the various effects of Sb substitution. Both the expansion of the unit-cell volume as well as electron doping will reduce the strength of the Kondo interaction J and thus further localize the $4f$ states as the Ce- $4f$ states are electron-like. Since Sb replaces Ge, this substitution results in a strongly disordered hybridization and therefore a distribution of the (local) Kondo temperatures: Sb substitution of the Ge atoms forming the cage enclosing the Ce leads to significant changes in the local environment of the Ce [27]. The increase in disorder is clearly reflected in the x -dependence of the residual resistivity (see Fig. 2). In addition, the relevant Kondo scale, $T_K \sim e^{-1/JN_F}$, is [at least for a structureless density of states (DOS)] also affected by the DOS at the Fermi level, N_F . Band structure calculations show (see Fig. 3a) that N_F shows an overall linear decrease with increasing x with a broad maximum around $x = 1.7$. A second maximum develops around $x = 4.2$ followed by an abrupt decrease of N_F down to 0 at $x \approx 5$ (not shown here), similar to the calculations done for the system $\text{LaPt}_4\text{Ge}_{12-x}\text{Sb}_x$ in a recent study [28]. These effects result in a strong decrease of the Kondo scale as x increases (which may happen to be somewhat slower around $x = 1.5$) and a broadening in the distribution of Kondo temperatures. The calculations based either on the experimental unit-cell volume expansion or taking the concentration-independent V_0 at $x = 0$ are nearly identical and agree well with a rigid-band approach. In this calculation, the Ce- $4f$ states are taken as core states. If Kondo correlations were negligible, the Sommerfeld coefficient should mirror the behavior of $N_F(x)$ of our band structure calculations. This is not the case (see Fig. 1 for $x \leq 1$), pointing to the effect of the Ce-moment localization as the Kondo scale decreases.

The appearance of the NFL regime is closely related to the non-magnetic disorder introduced by the Sb substitution and the resulting distribution in Kondo temperatures. Upon increasing x , the system evolves toward more localized f -moments as charge fluctuations decrease. The concomitant increase in disorder results in a distribution of the hybridization strength between the f -moments and conduction electrons. LFL develops at temperatures below the lowest Kondo scales of the distribution. The observed NFL behavior at $x = 1$ and 1.3 arises from a distribution of Kondo temperatures that extends below the lowest temperatures [29–31]. Such a scenario predicts a logarithmic divergence of $C_{\text{el}}(T)/T$

and a $\rho(T) = \rho_0 - aT$ dependence of the electrical resistivity, which we both find below 1 K in $\text{CePt}_4\text{Ge}_{11}\text{Sb}$ [see Fig. 1 for $C(T)/T$ and Fig. 2e for $\rho(T)$]. Within this scenario, applying a magnetic field removes the incoherent spin-flip scattering below some energy scale related to the magnetic field. This is reflected in the negative magnetoresistance near $x \approx 1$ (see Fig. 2e). Furthermore, the Kondo-disorder scenario gives a natural explanation for the extended Sb concentration range where the NFL behavior is observed.

As x increases further (beyond $x > 1.3$), the Kondo-temperature distribution continues to decrease in temperature. This lowering enhances the tendency toward magnetism. For $x = 1.5$, no long-range order develops but the maximum in resistivity indicates that scattering is suppressed below T_{max} . Since this maximum cannot be related to Kondo coherence, it is most likely caused by short-range order among the Ce moments. The resulting effective field induced by the short-range order suppresses incoherent spin-flip scattering below some scale, reminiscent of the effect of an external magnetic field (see Fig. 2e), thereby leading to a LFL groundstate. This is indeed observed. Finally, for $x \geq 1.9$, magnetic long-range order develops. Although the low temperature γ value of $\text{CePt}_4\text{Ge}_{10.1}\text{Sb}_{1.9}$ in the magnetically ordered state is still enhanced, it is much smaller than that at $x = 1.5$ as expected for weakened Kondo correlations. For even higher concentrations the estimated γ seems to decrease further. This substantiates that the Ce moments become even more localized with increasing substitution level and that the magnetism is of local rather than itinerant nature.

In summary, in $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$ the evolution of a local moment AF state starting from an intermediate-valent state is controlled by charge doping. While on increasing Sb concentration in $\text{CePt}_4\text{Ge}_{12-x}\text{Sb}_x$ local Ce moments begin to develop, they are at the same time becoming screened by the Kondo effect. The delicate relation of correlation effects together with a distribution in hybridization strength results in an unconventional phase diagram. The observation of a NFL regime in specific heat and electrical resistivity and the overall features of the phase diagram can be well explained by an interplay of correlation effects and Kondo disorder.

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[1] C. Uher, *Semiconductors and Semimetals* **69**, 139 (2001).
 [2] B. C. Sales, in *Handbook on the Physics and Chemistry of*

- Rare Earths, Vol. 33*, edited by K. A. Gschneidner Jr., J.-C. G. Bünzli, and V. K. Pecharsky (Elsevier, Amsterdam, 2003), chap. 211, pp. 1–34.
- [3] H. Sato, H. Sugawara, Y. Aoki, and H. Harima, in *Handbook of Magnetic Materials, Vol. 18*, edited by K. H. J. Buschow (Elsevier, Amsterdam, 2009), pp. 1–110.
- [4] M. B. Maple, Z. Henkie, R. E. Baumbach, T. A. Sayles, N. P. Butch, P.-C. Ho, T. Yanagisawa, W. M. Yuhasz, R. Wawryk, T. Cichorek, et al., *J. Phys. Soc. Jpn.* **77**, Suppl. A, 7 (2008).
- [5] E. Bauer, A. Grytsiv, X. Q. Chen, N. Melnychenko-Koblyuk, G. Hilscher, H. Kaldarar, H. Michor, E. Royanian, G. Giester, M. Rotter, et al., *Phys. Rev. Lett.* **99**, 217001 (2007).
- [6] R. Gumeniuk, W. Schnelle, H. Rosner, M. Nicklas, A. Leithe-Jasper, and Yu. Grin, *Phys. Rev. Lett.* **100**, 017002 (2008).
- [7] R. Gumeniuk, K. O. Kvashnina, W. Schnelle, M. Nicklas, H. Borrmann, H. Rosner, Y. Skourski, A. A. Tsirlin, A. Leithe-Jasper, and Yu. Grin, *J. Phys.: Condens. Matter* **23**, 465601 (2011).
- [8] R. Gumeniuk, M. Schöneich, A. Leithe-Jasper, W. Schnelle, M. Nicklas, H. Rosner, A. Ormeci, U. Burkhardt, M. Schmidt, U. Schwarz, et al., *New J. Phys.* **12**, 103035 (2010).
- [9] D. Kaczorowski and V. H. Tran, *Phys. Rev. B* **77**, 180504(R) (2008).
- [10] E. Bauer, X. Chen, P. Rogl, G. Hilscher, H. Michor, E. Royanian, R. Podlucky, G. Giester, O. Sologub, and A. P. Gonçalves, *Phys. Rev. B* **78**, 064516 (2008).
- [11] M. Nicklas, R. Gumeniuk, W. Schnelle, H. Rosner, A. Leithe-Jasper, F. Steglich, and Yu. Grin, *J. Phys.: Conf. Series* **273**, 012118 (2010).
- [12] H. Rosner, J. Gagner, D. Regesch, W. Schnelle, R. Gumeniuk, A. Leithe-Jasper, H. Fujiwara, T. Hauptrecht, T. C. Koethe, H. Hsieh, et al., *Phys. Rev. B* **80**, 075114 (2009).
- [13] R. Gumeniuk, H. Borrmann, A. Ormeci, H. Rosner, W. Schnelle, M. Nicklas, Yu. Grin, and A. Leithe-Jasper, *Z. Kristallogr.* **225**, 531 (2010).
- [14] R. Gumeniuk, H. Rosner, W. Schnelle, M. Nicklas, A. Leithe-Jasper, and Yu. Grin, *Phys. Rev. B* **78**, 052504 (2008).
- [15] M. Toda, H. Sugawara, K.-I. Magishi, T. Saito, K. Koyama, Y. Aoki, and H. Sato, *J. Phys. Soc. Japan* **77**, 124702 (2008).
- [16] A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, 1993).
- [17] H. v. Löhneysen, A. Rosch, M. Vojta, and P. Wölfle, *Rev. Mod. Phys.* **79**, 1015 (2007).
- [18] Q. Si and F. Steglich, *Science* **329**, 1161 (2010).
- [19] Y. Matsumoto, S. Nakatsuji, K. Kuga, Y. Karaki, N. Horie, Y. Shimura, T. Sakakibara, A. H. Nevidomskyy, and P. Coleman, *Science* **331**, 316 (2011).
- [20] J. Lawrence, *Phys. Rev. B* **20**, 3770 (1979).
- [21] P. Pedrazzini, M. G. Berisso, N. Caroca-Canales, M. Deppe, C. Geibel, and J. G. Sereni, *Eur. Phys. J. B* **38**, 445 (2004).
- [22] R. Gumeniuk *et al.*, unpublished.
- [23] The substitution of Ge by As is difficult and results in multi-phase samples.
- [24] M. Nicklas *et al.*, *Simultaneous suppression of antiferromagnetic order and Kondo coherence in CePt₄Ge_{12-x}Sb_x*, SCES11 conference contribution (2011), cambridge, UK.
- [25] K. Koepernik and H. Eschrig, *Phys. Rev. B* **59**, 1743 (1999).
- [26] J. Perdew and Y. Wang, *Phys. Rev. B* **45**, 13244 (1992).
- [27] Note, that the Γ_7 doublet groundstate is in line with expectations for a $J = 5/2$ state in a cubic environment.
- [28] M. X. Chen, R. Podlucky, S. Humer, E. Bauer, and P. Rogl (2012), arXiv:1206.4428.
- [29] V. Dobrosavljevic, T. R. Kirkpatrick, and G. Kotliar, *Phys. Rev. Lett.* **69**, 1113 (1992).
- [30] E. Miranda, V. Dobrosavljevic, and G. Kotliar, *J. Phys.: Condens. Matter* **8**, 9871 (1996).
- [31] E. Miranda, V. Dobrosavljevic, and G. Kotliar, *Phys. Rev. Lett.* **78**, 290 (1997).