Fluctuation-induced first-order transition in Eu-based trillium lattices

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Among spin arrangements prone to geometric frustration, the so-called trillium lattice has not been very intensively investigated. A few theoretical works show that it is at the border between a degenerate, an only partially ordered, and a fully ordered ground state. However, only few compounds with this structure have been studied, and there is presently no good example of a trillium lattice with an antiferromagnetic ground state and clear evidence for frustration effects. We present magnetic and specific heat measurements on two realizations of a trillium lattice of local spins, EuPtSi and EuPtGe. Both compounds exhibit a similar magnetic behavior, with Eu^{2+} moments ordering antiferromagnetically at T_N=4.1 K (EuPtSi) and 3.3 K (EuPtGe), albeit retaining a considerable amount of entropy in strong magnetic fluctuations extending to temperatures well above T_N. The magnetic entropy reaches only roughly half of $R \ln 8$ at T_N. These fluctuations are presumably the source for the pronounced first-order character of the transition at T_N and are likely due to magnetic frustration. Thus, EuPtSi and EuPtGe open a new door to experimental studies of frustration effects in the trillium lattice and provide a testing ground for theoretical predictions.

I. INTRODUCTION

Geometric frustration in spin lattices leads to unconventional ground states and exotic low-energy excitations. The fundamental characteristic of a frustrated ground state is its large degeneracy, which, following Moessner and Chalker [1], can be roughly estimated as a function of the number of spin components, the number of spins within a unit (usually a triangle or tetrahedron), and the connectivity of the units (i.e., how many units share a spin). The trillium lattice [2] plotted in Fig. 1, a three-dimensional network of corner-sharing triangles resembling a trillium flower, appears to be at the border between a frustrated degenerated ground state and an ordered one. For the classical Heisenberg model, assuming a "hard" spin constraint, $|s_i|^2 = 1$ leads to a nondegenerate ground state, but relaxing the constraint to $\Sigma |s_i^\alpha|^2 = 1$ (where the summation is over the trillium sites within a unit cell of the global lattice) yields, in the large-N limit, a degenerate ground state [2] with a continuum of **q** vectors on a spheroid centered at $\mathbf{q} = (0,0,0)$ called partial order. Monte Carlo simulations for the "soft" constraint predict a strong first-order transition to the ordered state, with prominent partial-order fluctuations above the ordering temperature [3]. Considering Ising spins with a local anisotropy and ferromagnetic interactions gives rise to a spin-ice–like ground state [4,5].

In materials crystallizing with space group $P2_1 3$, the atoms located at $4a$ sites (site symmetry .3.) form a trillium lattice (see Fig. 1). The best-known examples are B20 compounds like MnSi (FeSi structure type) [6]. However, these materials are essentially ferromagnets, where the Dzyaloshinskii-Moriya interaction induces a spiral magnetic structure with very small propagation vectors **q** [7,8]. Under certain conditions a magnetic field can induce magnetic skyrmions, topologically stable particlelike objects, which have attracted considerable

interest [9,10]. Because of the strong ferromagnetic interaction, frustration does not appear to be significant in these systems. Further realizations of the trillium lattice include intermetallic compounds EuPtSi and EuPtGe (LaIrSi structure type). Early works on these compounds [11,12] did not observe a clear signature of magnetic order in the magnetic susceptibility or the specific heat, in spite of Curie-Weiss temperatures of 5 K for EuPtSi and 20 K for EuPtGe. In EuPtSi a significant broadening of the Mössbauer line at 4.2 K merely evidenced the onset of magnetic order at this temperature [11], while no broadening was observed in EuPtGe [12] at 4.2 K. In this work, we show that both compounds present an unusual magnetic behavior. They ultimately order antiferromagnetically at T_N=4.1 K (EuPtSi) and 3.3 K (EuPtGe), but a shoulder in the magnetic susceptibility above T_N and a strongly reduced entropy at T_N provide clear evidence for strong antiferromagnetic fluctuations extending far above T_N. Furthermore, the transition at T_N has a very pronounced first-order character, which is likely a consequence of large fluctuations. These results indicate frustration to be an important ingredient in the magnetism of EuPtSi and EuPtGe.

II. EXPERIMENTAL

Polycrystals of EuPtSi and EuPtGe were synthesized from high-purity Eu (Ames Laboratory), Pt wire (Heraeus, 99.99%), and Si (Alfa Aesar, 99.9999%) or Ge (Alfa Aesar, 99.9999+%) pieces. Platinum and silicon or germanium were prereacted in an arc furnace in order to decrease the melting temperature (1232° for PtSi and 1070° for PtGe, compared to 1770° for Pt) below the boiling temperature of Eu (1527 °). In the case of EuPtSi, the resulting material was coarsely crushed and arc-melted together with Eu. The initial weight loss was compensated by subsequent addition of Eu and remelting. In the case of EuPtGe, better sample quality was achieved by heating PtGe and Eu in a sealed tantalum crucible up to 1160 °C. Postannealing at 700° did not improve the quality
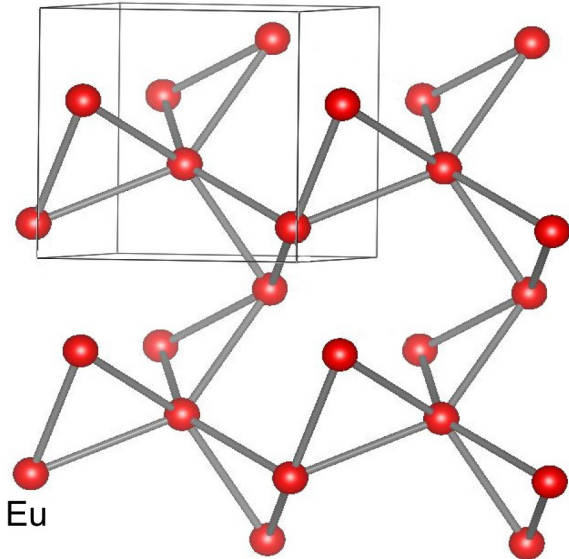
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FIG. 1. Trillium lattice formed by Eu atoms in EuPtSi and EuPtGe. The cubic unit cell is indicated.

of the samples; therefore only results for as-cast samples are shown here.

The synthesized samples were characterized by powder x-ray diffraction: laboratory (Cu $K\alpha$ radiation) and synchrotron (ID22 beamline of ESRF, LaB$_6$ as internal standard, $\lambda = 0.400\,14(4)$ Å). The crystal structure of EuPtSi was refined using single-crystal data (Rigaku AFC7 diffractometer, Saturn 724+ CCD detector, Mo $K\alpha$ radiation). The chemical composition was characterized by microprobe analysis with energy-dispersive x-ray spectroscopy (EDX). Heat capacity measurements were performed in a Quantum Design PPMS system with ^{3}He option. Magnetization data were taken using a Quantum Design MPMS superconducting quantum interference device (SQUID) magnetometer.

III. RESULTS AND DISCUSSION

X-ray powder diffraction confirmed the cubic LaIrSi-type structure (space group $P2_13$) for all samples. Lattice parameters are in agreement with previous reports [11,12]. While EuPtGe samples were found to be single-phased, in EuPtSi samples a few micron-sized inclusions of secondary phase were observed, consistent with a small amount of EuPt$_2$Si$_2$ (CaBe$_2$Ge$_2$-type) detected in powder diffraction. Structural refinement for EuPtSi is summarized in Table I. For the calculation of interatomic distances, lattice parameters obtained from powder synchrotron data were used. To check the composition, in a separate run the occupancies of all atomic positions were refined as free variables. The obtained values [1.00(2) for all sites] confirmed a 1:1:1 stoichiometry. The equiatomic composition was also corroborated by microprobe analysis.

As previously reported [11,12], the magnetic susceptibility at high temperatures (not shown) is well described by a Curie-Weiss law in both compounds. The effective moment deduced from the slope of the inverse susceptibility versus temperature

TABLE I. Crystal structure data of EuPtSi [space group $P2_13$, $a = 6.436\,11(1)$ Å]. [a]

Atom	Wyckoff site	x	U_{eq}
Eu	$4a$ (x,x,x)	0.37001(4)	0.0111(1)
Pt	$4a$ (x,x,x)	0.08630(3)	0.00959(9)
Si	$4a$ (x,x,x)	0.6611(2)	0.0103(7)

[a]The refinement of the structure (LaIrSi structure type) revealed residuals R1 = 0.0198 and wR2 = 0.030 (590 and 595 observed reflections, respectively).

in the range 100–380 K is close to that expected for Eu^{2+} in both samples, while the intercept results in slightly positive (ferromagnetic) Curie-Weiss temperatures, $\Theta_W = 9.8$ K (EuPtSi) and 4.6 K (EuPtGe). However, a precise analysis of the inverse susceptibility as a function of temperature reveals a slightly positive curvature. This curvature might be due to the influence of a small amount of foreign phase, and/or the effect of the onset of antiferromagnetic correlations. The presence of a tiny amount (of the order of 0.5%) of impurity phase, likely EuO, is evidenced by a ferromagnetic anomaly visible in the small field susceptibility (see Appendix). Due to this slight positive curvature, decreasing the temperature range of the Curie-Weiss fit results in a decrease of the obtained Θ_W, though it remains positive. Θ_W therefore cannot be precisely determined, but it is positive for both compounds, with an upper limit given by the high temperature fits. Thus, exchange interactions are predominantly ferromagnetic, though the small values of Θ_W in connection with other properties suggests a competition between ferromagnetic and antiferromagnetic interactions. Below 30 K the positive curvature in the inverse susceptibility becomes pronounced, indicating the onset of antiferromagnetic correlations far above T_N.

A sharp drop in the susceptibility at 4.2 K for EuPtSi and 3.2 K for EuPtGe marks the onset of antiferromagnetic order. In view of the ferromagnetic Θ_W, the chiral crystal structure, and the known magnetic structure in the intensively studied MnSi-type systems, one might suspect the magnetic state in the Eu systems to be a nearly ferromagnetic, small **q** spiral as in the MnSi-type compounds. However, magnetization measurements at $T = 2$ K $< T_N$ in the inset of Fig. 2(a) show an almost linear increase in magnetization versus applied field up to ~2.5 T (EuPtSi) and 3 T (EuPtGe), with saturation at slightly larger fields. The saturation moment is close to the expected $\mu_{sat} = 7\mu_B$ for Eu^{2+}. The large field range over which the magnetization increases linearly indicates a true antiferromagnetic ordering, with the saturation field providing an order of magnitude for the strength of antiferromagnetic interactions. Just above T_N, the susceptibility slowly decreases upon warming with a temperature dependence much weaker than a Curie-Weiss behavior, further confirming the presence of strong antiferromagnetic correlations above T_N(see also Fig. 5). In the case of EuPtGe, a second anomaly is observed at 2.6 K. The small irreversibility observed between field-cooled and zero-field-cooled runs stems most probably from the ferromagnetic impurity phase.
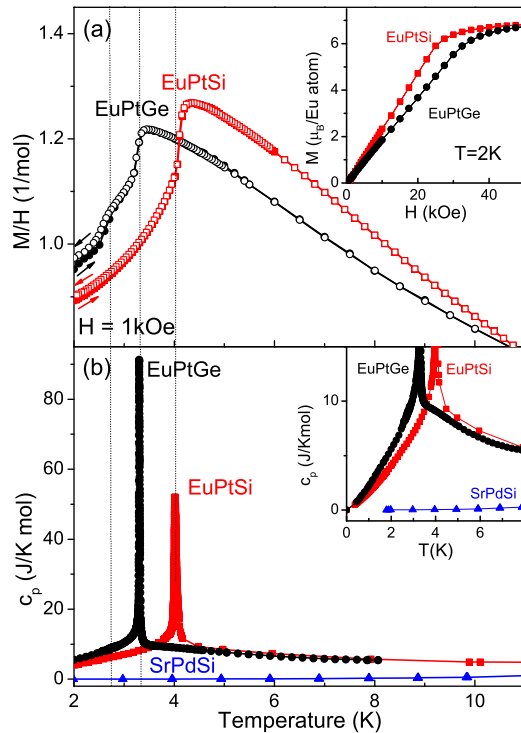
FIG. 3. Specific heat data in the vicinity of T_N for EuPtGe (left) and EuPtSi (right) obtained from the application of a single heat pulse (full symbols) and upon the subsequent cooling (open symbols). The actual evolution of sample temperature with time is shown in the insets; the transition temperatures can be clearly identified both upon warming and cooling.

FIG. 2. (a) Magnetic susceptibility of EuPtSi (red squares) and EuPtGe (black circles) as a function of temperature measured at an applied field of 1 KOe. Full symbols represent data taken after zero-field cooling and open symbols correspond to field-cooling data. Inset: Magnetization vs applied field for EuPtSi (red squares) and EuPtGe (black circles). (b) Specific heat of EuPtSi (red squares) and EuPtGe (black circles) measured as a function of temperature, compared to the nonmagnetic isostructural compound SrPdSi (blue triangles). Inset: Low-temperature specific heat data. Specific heat data in the vicinity of the transition have been obtained from a single heat pulse; data away from the transition were obtained by the standard analysis.

Specific heat measurements as a function of temperature in Fig. 2(b) underline the bulk character of the antiferromagnetic transition seen in the magnetic susceptibility for both EuPtSi and EuPtGe. An extremely narrow but very high specific heat peak is observed at 4.1 K for EuPtSi and 3.3 K for EuPtGe, while no clear anomaly was detected in EuPtGe at 2.6 K. The height of the peaks at T_N is much larger than the expected specific heat jump within a mean-field approximation [13]: 20.15 J/K mol for 7/2 spins such as Eu and Gd. This fact, as well as the sharpness of the anomaly, suggests that the magnetic transition is first order. Notice that close to the magnetic transition the specific heat changes rapidly with temperature. The heat capacity option of PPMS extracts a specific heat value by globally fitting a temperature vs time relaxation curve as a heating pulse is applied and switched off, assuming a constant sample specific heat for the temperature range involved. (It can also take into account a finite thermal conductance between the sample and the sample platform in the so-called "two-tau" scheme [14].) In the case of a strongly temperature-dependent specific heat, as in a first-order transition, this procedure is inadequate [15]. Specific heat values in the vicinity of the transition were then obtained from a single slope analys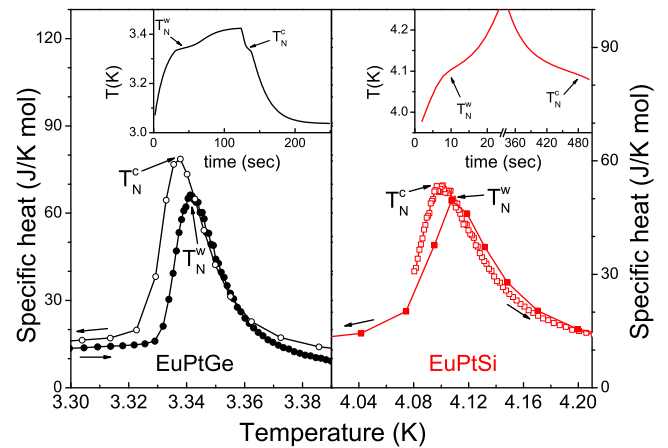is of relaxation curves. For this, a small range of the relaxation curve is used to obtain the specific heat at that particular temperature range under the assumption of an infinite thermal conductivity between platform and sample ("single tau") [14,15].

The first-order character of the transition is highlighted by the time dependence of the sample temperature upon application of a heat pulse at a temperature just below the transition (see inset of Fig. 3). The presence of the first-order transition is clearly observed in the dramatic change of slope of temperature vs time, both during warming and cooling. The anomaly was observed at a few millikelvin lower temperature upon cooling than upon warming. Both the transition temperature and the shift between warming and cooling were found to be dependent (within ∼10 mK) on the heater power, as was the height of the computed specific heat peak. The anomaly at T_N is superimposed on a large broad peak characteristic of magnetic fluctuations. The general shape of the specific heat curve shows some similarity to that for layered magnetic systems [16–18], where a specific heat peak corresponding to three-dimensional order develops on top of a broad two-dimensional contribution. The cubic structure of EuPtSi and EuPtGe and the three-dimensional nature of the magnetic lattice, however, suggest rather three-dimensional fluctuations. In any case, magnetic fluctuations in EuPtSi and EuPtGe extend up to temperatures at least several times the ordering temperature.

This is substantiated by an analysis of the magnetic entropy $S_{\mathrm{mag}}(T)$ (see Fig. 4). In order to compute $S_{\mathrm{mag}}(T)$, the contributions of phonons and conduction electrons to the specific heat need to be subtracted. Comparing the specific heat of EuPtSi and EuPtGe with that of an isostructural isoelectronic nonmagnetic reference compound, SrPdSi, shows that below 10 K the phonon contribution becomes much smaller than the magnetic one [see Fig. 2(b)]. The magnetic contribution was estimated by simply subtracting the specific heat of
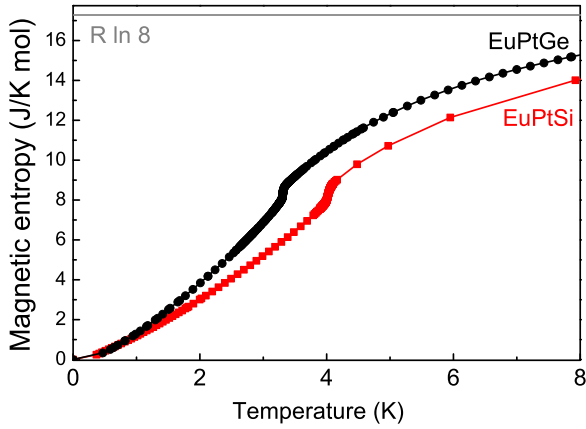
FIG. 4. Magnetic entropy computed from the specific heat data after subtraction of the specific heat of SrPdSi. The entropy corresponding to the full degeneracy of the 7/2 moment of Eu^{2+}, R ln 8, is indicated for comparison.

SrPdSi from that of EuPtSi and EuPdGe.[1] Due to the higher atomic masses involved, Debye temperatures should be lower and correspondingly, the phonon contribution higher in the Eu-based compounds. Ignoring the larger masses of the latter results in an overestimation of the magnetic specific heat at high temperatures, leading to a slight overestimation of $S_{mag}(T)$. The specific heat contribution from conduction electrons was assumed to be the same in all compounds. An eventual 10-mJ/K^2 mol difference between the Sommerfeld coefficient of magnetic and nonmagnetic compounds would imply a change in $S_{mag}(10 K)$ of merely 0.01 R, which can safely be ignored in the further analysis. In spite of the fact that our approximation leads to a slight overestimation of S_{mag} at high temperatures, the magnetic entropy at T_N is roughly only half the full R ln8 expected for the 7/2 spin of Eu^{2+}. Part of the missing magnetic entropy at T_N could stem from an underestimation of the specific heat peak height due to the experimental method, but this cannot be a large effect since at much higher temperatures we overestimate S_{mag}. Furthermore, the large tail of the specific heat curve in the inset of Fig. 2(b) provides strong evidence that a substantial amount of entropy is carried by the magnetic fluctuations above the Néel temperature. The estimated entropy change at the transition is $\Delta S = 0.5$ J/K mol for EuPtGe and 0.75 J/K mol for EuPtSi, yielding an enthalpy change or latent heat $\Delta H = T \Delta S = 1.7$ and 3 J/mol, respectively. These values are little influenced by the choice of phonon and electronic background but possibly represent a lower boundary, since they are linked to the measured height of the specific heat peak.

Such pronounced first-order-type magnetic transitions are not common in Eu^{2+}- or Gd^{3+}-based systems. The absence of orbital moment in the $J = S = 7/2$ pure spin ground-state multiplet of Eu^{2+} and Gd^{3+} prevents the strong magnetocrystalline anisotropy common to many rare-earth systems, which through magnetoelastic coupling is a frequent cause for driving the magnetic transition first order. However, under certain conditions, in an isotropic or weakly anisotropic system a second-order transition can be driven first order by strong fluctuations [19]. An example is the helimagnetic-paramagnetic transition in the structurally related compound MnSi [20]. There, the Dzyaloshinskii-Moriya (DM) interactions due to the absence of inversion symmetry play a crucial role for inducing strong fluctuations. In the present Eu-based compounds, despite the absence of orbital moment, a DM interaction is likely present because Pt $5d$ states, subject to a strong spin-orbit interaction, are expected to play a significant role in the Ruderman-Kittel-Kasuya-Yosida type of exchange between Eu moments. However, the present results on EuPtSi and EuPtGe suggest that in these compounds magnetic frustration emerging from the geometrical frustration inherent to the trillium geometry of the lattice and/or from competing ferromagnetic and antiferromagnetic interactions is a more important source of fluctuations, in addition to the DM interaction. Note that although these systems likely present true antiferromagnetic order, recent studies demonstrated the possibility of skyrmion phases in frustrated antiferromagnetic systems [21,22].

IV. CONCLUSIONS

We show that two realizations of the trillium lattice of local Eu^{2+} moments, EuPtSi and EuPtGe, present unusual magnetic behavior. Upon cooling, long-range antiferromagnetic order at T_N=4.1 K (EuPtSi) and 3.3 K (EuPtGe) develops from a background of strong magnetic fluctuations extending to temperatures well above T_N. The extent of the fluctuations is highlighted by the magnetic entropy, that reaches only half of R ln8 at T_N. The transitions at the respective T_N's present a strong first-order character, which is likely a consequence of the large fluctuations. Magnetic frustration, which is probably at the origin of these large fluctuations, might be due to the geometry of the trillium lattice, to a competition between ferro- and antiferromagnetic exchange, or to a Dzyaloshinskii-Moriya interaction caused by the combination of a noncentrosymmetric structure and Ruderman-Kittel-Kasuya-Yosida exchange mediated by the heavy Pt atoms. Since metallic frustrated local moment systems are rare, EuPtSi and EuPtGe provide a new experimental testing ground and motivate the further development of theoretical studies of frustration effects in the trillium lattice.

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[1]Due to the smaller atomic masses, the Debye temperature of SrPdSi, Θ_{SrPdSi}, is expected to be higher than that of EuPtSi and EuPtGe. Estimations of the phonon background from the SrPdSi data using a temperature renormalization by $\Theta_{EuPtX}/\Theta_{SrPdSi}$ were unreasonably large, using both $\Theta_{EuPtX}/\Theta_{SrPdSi} = (M_{SrPdSi}/M_{EuPtX})^{1/2}$ and $\Theta_{EuPtX}/\Theta_{SrPdSi} = [(M_{Sr}^{3/2} + M_{Pd}^{3/2} + M_{Si}^{3/2})/(M_{Eu}^{3/2} + M_{Pt}^{3/2} + M_{X}^{3/2})]^{1/3}$ schemes, resulting in negative magnetic specific heat at high temperatures.
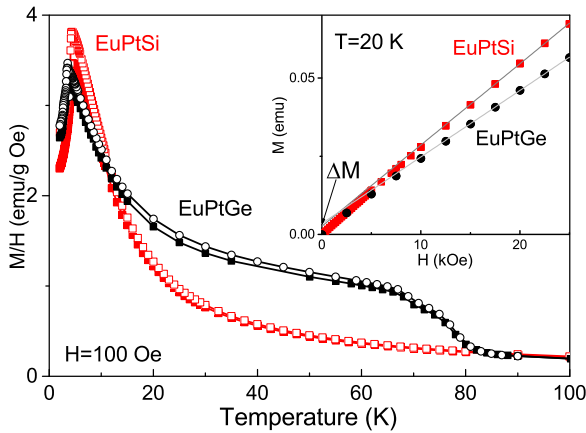
FIG. 5. Magnetic susceptibility of EuPtSi (red squares) and EuPtGe samples (black circles) at 100 Oe. Open symbols represent data taken on field cooling; full symbols were taken upon warming from a zero-field-cooled state. Inset: Magnetization curve taken/ at 20 K.

APPENDIX: FERROMAGNETIC IMPURITY PHASE

Previous works on these materials reported the presence of a magnetic impurity phase [11,12], undetected in powder diffraction patterns but clearly visible as an increase in the magnetic susceptibility or an additional specific heat contribution at temperatures in the range 70–80 K. This impurity is presumably EuO, which for the stoichiometric composition orders ferromagnetically at $T_C = 69$ K [23] but can reach $T_C \approx 150$ K upon oxygen deficiency [24]. Due to the high reactivity of Eu metal with air and moisture, the formation of the phase is hardly surprising. In our EuPtGe samples, the magnetic susceptibility at low fields (100 Oe) shows a clear signature of the ferromagnetic transition (see Fig. 5). In our EuPtSi samples the ferromagnetic transition is not detected at T_C, but a small irreversibility between field-cooled and zero-field-cooled runs suggests ferromagnetic impurities are nonetheless present. At temperatures well below T_C but much higher than the ordering temperature of the 111 phase, for example, at 20 K, it may be assumed that at moderately large fields (e.g., around 20 kOe) the EuO moments are fully saturated, while the magnetic response of the 111 phase is linear to the applied field. The inset of Fig. 5 shows magnetization curves for our EuPtSi and EuPtGe samples at 20 K. By performing a linear fit of the data in the range 15–25 kOe, it is possible to estimate the intrinsic paramagnetic response of the 111 phase from the slope, while the intercept yields the excess magnetization ΔM due to the contribution of (magnetically saturated) EuO. At higher fields, the slope of $M(H)$ decreases due to the beginning of saturation of the intrinsic magnetization. The mass fraction f of the EuO impurity is $f = \frac{\Delta M}{N_A g J \mu_B} \frac{m_{mol}^{EuO}}{m_{tot}}$, where $N_A g J \mu_B$ is the magnetization per mol of Eu^{2+}, m_{mol}^{EuO} the molar mass of EuO, and m_{tot} the total sample mass. In the case of EuPtGe, the mass fraction of the impurity phase thus obtained is 0.6%, while for EuPtSi it is 0.48%. Note that estimations of the EuO content from the magnetization step at T_C in small fields can severely underestimate the EuO fraction.

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