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# Pronounced first-order metamagnetic transition in the paramagnetic heavy-fermion system CeTiGe

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We report on the observation of large, steplike anomalies in the magnetization ($\Delta M = 0.74\ \mu_B$/Ce), in the magnetostriction ($\Delta l/l_0 = 2.0 \times 10^{-3}$), and in the magnetoresistance in polycrystals of the paramagnetic heavy-fermion system CeTiGe at a critical magnetic field $\mu_0 H_c \approx 12$ T at low temperatures. The size of these anomalies is much larger than those reported for the prototypical heavy-fermion metamagnet $CeRu_2Si_2$. Furthermore, hysteresis between increasing and decreasing field data indicate a real thermodynamic, first-order type of phase transition, in contrast to the crossover reported for $CeRu_2Si_2$. Analysis of the resistivity data shows a pronounced decrease of the electronic quasiparticle mass across $H_c$. These results establish CeTiGe as a rare metamagnetic Kondo-lattice system, with an exceptionally large, metamagnetic transition of first-order type at a moderate field.

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Kondo lattices (KLs) are intermetallic compounds based typically on a rare-earth element with an unstable *f* shell, usually Ce or Yb. Because of this instability they can be tuned from a magnetic to a nonmagnetic state by applying mechanical or chemical pressure. At the crossover, one observes very unusual properties, e.g., the formation of heavy quasiparticles (heavy fermions), unconventional superconductivity, and non-Fermi-liquid (nFL) ground states.[1,2] Accordingly, these systems have been the subject of continuous intensive research for more than 30 years.

Most recently, the fate of the 4*f* electrons, i.e., how the related degrees of freedom are removed from the Fermi surface through localization of the *f* electrons, has developed into a central issue in the field.[1,2] In this context, another unusual phenomenon in KLs has recently regained considerable attention: the pronounced metamagnetic transition (MMT) observed in the paramagnetic heavy-fermion (HF) system $CeRu_2Si_2$.[3] At a critical field $H_c = 7.7$ T applied along the tetragonal *c* axis, which is the easy axis for this Ising-type system, one observes a large step in the field-dependent magnetization $M(H)$ of the order of 0.4 $\mu_B$/Ce. Very accurate measurements down to lowest temperatures indicate that this transition is not a real thermodynamic phase transition, but rather a sharp crossover, since the transition width and height remain finite down to the lowest temperatures.[4] Despite intensive studies over the past 20 years, the nature of this transition is still a matter of discussion. Strong changes of the Fermi surface at $H_c$ were taken as evidence for a transition from an itinerant $4f$ at $H < H_c$ to a localized 4*f* state at $H > H_c$.[5] On the contrary, a recent study of the magnetoresistance suggests that this transition corresponds to a continuous disappearance of one of the spin-split sheets of the Fermi surface.[6] One of the problems for a more general study of this type of MMT is the absence of further good examples in $4f$-based Kondo lattices at accessible fields. Metamagnetic transitions at low fields have been recently reported for the Yb-based HF systems $YbT_2Zn_{20}$ ($T$ = Co, Rh, Ir), but they are very broad, pointing to a crossover.[7] Magnetization curves with a sharp step in $M(H)$ have been reported for $CeNi_2Ge_2$ (Ref. 8) and $CeFe_2Ge_2$.[9] However, the critical fields are much larger (42 and 30 T, respectively), and the steps in $M(H)$ are significantly smaller ($\leqslant 0.2\ \mu_B$/Ce). Therefore, the MMTs in these systems have not been deeply investigated. In literature many other KLs have been proposed to exhibit a MMT, such as, e.g., $CeCu_6$ (Ref. 10) or $YbRh_2Si_2$,[11] but these compounds show only a kink in $M(H)$, not a step. This means that some essential ingredients are missing in those systems, which are present in $CeRu_2Si_2$.

Here, we report the discovery of a unique example of a paramagnetic KL which shows a pronounced MMT: CeTiGe. Our recent comprehensive study of the physical properties of CeTiGe established this compound as a different paramagnetic HF system with a Sommerfeld coefficient $\gamma \approx 0.3\ \mathrm{J/K^2\,mol}$,[12] being located on the nonmagnetic side of the quantum critical point (QCP) within the Doniach phase diagram. A maximum in the susceptibility $\chi(T)$ at 25 K and in the 4*f* specific heat $C_{4f}(T)/T$ at 16 K, as well as a single maximum in the thermopower $S(T)$ at 17 K (with a huge peak value of 60 $\mu$V/K), indicate a large degeneracy of the local moment involved. Both $\chi(T)$ and $C_{4f}(T)$ could be well fitted with the Coqblin-Schrieffer model for $J = 5/2$ and $T_0 = 82$ K, corresponding to a Kondo scale $T_K = 55$ K.[12] Preliminary results, indicating the presence of a MMT, have been reported in Ref. 13. The findings presented here are (i) a large step in the magnetization $\Delta M(H) = 0.74\ \mu_B$/Ce at a quite low field of about 12.5 T, which is accessible with a standard superconducting magnet, (ii) the MMT is accompanied by a hysteresis in magnetization, magnetoresistance, and magnetostriction loops, which strongly suggests the presence of a real thermodynamic first-order phase transition instead of a crossover as reported for $CeRu_2Si_2$,[14] and (iii) the analysis of the resistivity data shows an abrupt decrease of the electronic quasiparticle mass across the MMT. Hence, our discovery opens a door for the study of this unusual phenomenon.

Several samples were prepared with slight differences in the annealing temperature which ranged from 1000 up to 1150 °C.[12] Furthermore, we prepared few samples where a small amount of Ti or Ge was substituted by Zr, Nb, or Si, respectively. All samples showed the huge anomalies marking the MMT. Therefore, this transition is a robust, intrinsic property of CeTiGe, as long as it crystallizes in the

CeFeSi-structure type. Recently, Chevalier *et al.* reported the observation of a slightly different structure (CeScSi type) in quenched, off-stoichiometric CeTiGe powder samples.[15] We did not found any evidence of the presence of this phase in our samples, and further experiments under way suggest this structure type to be connected with off stoichiometry. The magnetoresistance and the linear magnetostriction were measured between 0.02 and 4 K in fields up to 18 T in a $^3$He/$^4$He dilution refrigerator using a standard four-terminal ac technique and a high-resolution CuBe capacitance dilatometer. The magnetization data were collected on powdered samples in a pulsed 60-T magnet down to 1.4 K at the Dresden High Magnetic Field Laboratory[16] and scaled by values taken in a superconducting quantum interference device (SQUID) magnetometer.

The essential result is displayed by the magnetization shown in Fig. 1 as $M(H)$ vs $H$ for $T = 1.4$ and 4.3 K. The prominent feature is a steplike anomaly at a critical field of $\mu_0 H_c \approx 12$ T. At lower and higher fields $M(H)$ increases weakly and almost linearly with $H$. The value at the highest field is slightly lower than the saturation magnetization expected for a fully localized Ce$^{3+}$ (2.14 $\mu_B$/Ce). Extrapolating the linear $M(H)$ toward $H_c$, an idealized magnetization step size of $\Delta M = 0.74\ \mu_B$/Ce at 1.4 K can be extracted. This is almost twice the value reported for CeRu$_2$Si$_2$, and a factor of 4 larger than in CeNi$_2$Ge$_2$ (Ref. 8) or in CeFe$_2$Ge$_2$.[9] Magnetization data taken on increasing and decreasing magnetic field show a hysteresis loop. This is better seen in the differential susceptibility $\partial M/\partial H$ displayed in the inset of Fig. 1 for the field range $6 \leqslant \mu_0 H \leqslant 19$ T: A sharp peak at 12.5 T for increasing field and one at 11.7 T for decreasing field yield a MMT with a hysteresis width of $\Delta\mu_0 H \approx 0.8$ T, inferring a transition of the first-order type. The difference in peak height and width between increasing and decreasing $H$ is likely related to the asymmetric field pulse which presents a steeper rising edge,[16] and to magnetocaloric effects. The peak positions are identical for 1.4 and 4.3 K, indicating a vertical phase boundary in the $H$-$T$ phase diagram. Therefore, the hysteresis cannot result from a shift in $T$ during the measurement. The $M(H)$ values at 4.3 K are slightly lower than those at 1.4 K, but the difference is within the absolute accuracy of the measurement. A small splitting opens below 47 T in the data at 1.4 K, which might be related to the magnetocaloric effect or to the alignment of the powder at high field, since the data for 1.4 K were taken first.

The short time scale of pulsed field experiments induces nonequilibrium effects, e.g., temperature shifts due to the magnetocaloric effect. For this reason we focused on experiments at very low temperatures and in static magnetic fields. Figure 2(a) shows the magnetoresistance between 6 and 18 T (sweeping rate of 0.5 mT/s) at temperatures in the range $0.02 \leqslant T \leqslant 4$ K. In order to minimize the effects due to the Lorentz force we have probed the longitudinal magnetoresistance with the current density $j \parallel H$. The most prominent feature in Fig. 2(a) is a sharp drop of the resistivity at $\mu_0 H_c \approx 12$ T with a pronounced hysteresis opening between the up and down field sweeps. The derivative $\partial\rho/\partial H$, plotted for the field range $7 \leqslant \mu_0 H \leqslant 18$ T in Fig. 2(b), emphasizes the sharpness of the transition by showing striking peaks at 12.9 T for increasing field and at 11.5 T for decreasing field, reminiscent of those observed in $\partial M(H)/\partial H$. Thus, the mean value of the critical field $\mu_0 H_c = 12.2$ T is the same as in $M(H)$, while the hysteresis is even larger. While the peak positions do not significantly shift with temperature, as for the magnetization, the peak size, i.e., the size of the drop in $\rho(T)$, increases strongly with increasing $T$, in contrast to the behavior of $M(H)$. However, this increase can be explained straightforwardly: It is directly connected with the strong increase of the zero-field resistivity with temperature, while the resistivity measured at fields larger than $H_c$ increases only very weakly with $T$. This effect deserves a deeper analysis, since it gives further insight into the nature of the MMT. In Fig. 3(a) the temperature dependence of the resistivity is plotted as $\rho(T)$ vs $T^2$ for selected fields

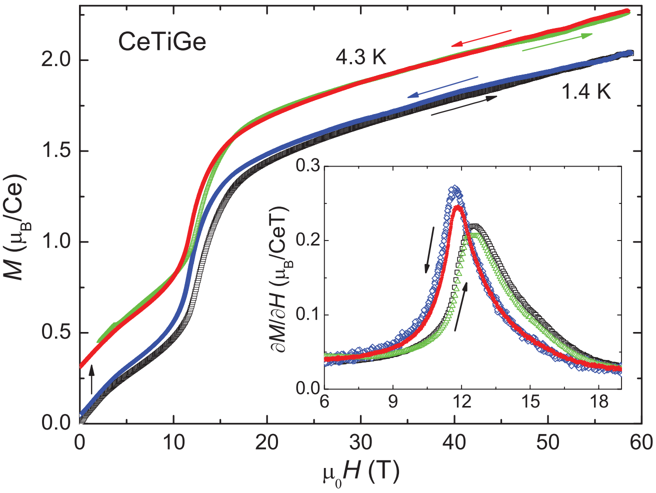

FIG. 1. (Color online) Magnetization $M(H)$ in fields up to $\mu_0 H = 60$ T measured at 1.4 and 4.3 K. The data at 4.3 K were shifted upward by 0.25 $\mu_B$/Ce. The arrows indicate the field direction. A step with $\Delta M = 0.74\ \mu_B$/Ce is observed at $\mu_0 H_c = (12.1 \pm 0.4)$ T. The inset shows the derivative $\partial M/\partial H$ vs $H$ for $6 < \mu_0 H < 19$ T and both temperatures.

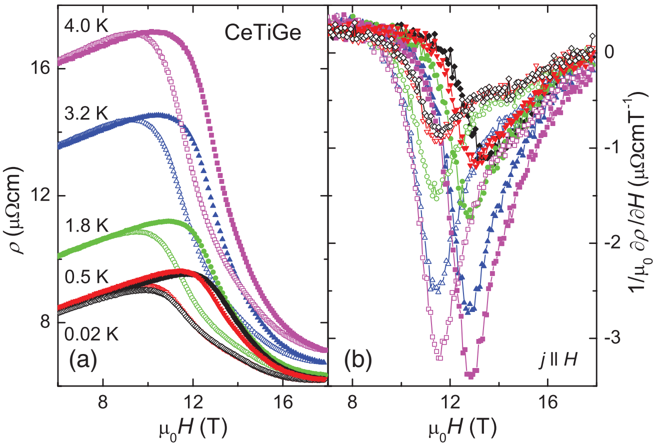

FIG. 2. (Color online) (a) Magnetoresistance $\rho(H)$ of polycrystals with current $j \parallel H$ measured at different temperatures between 0.02 and 4 K. A hysteresis loop between data collected with increasing field (solid symbols) and those with decreasing field (open symbols) is observed for all $T$. (b) The derivative $\partial\rho/\partial H$ is plotted for $7 \leqslant \mu_0 H \leqslant 18$ T.

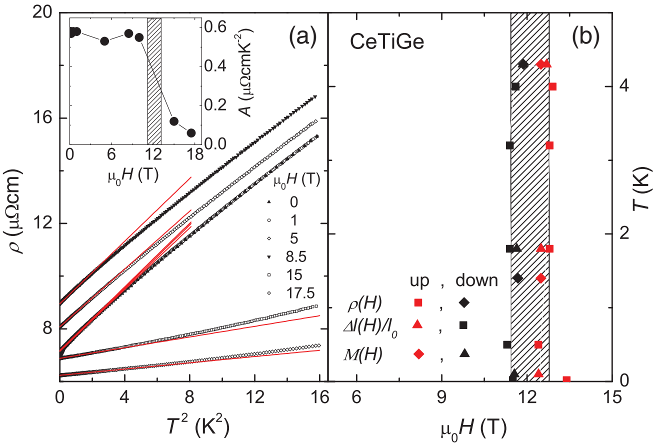

FIG. 3. (Color online) (a) Electrical resistivity $\rho(T)$ vs $T^2$ for different fields up to 17.5 T. Symbols indicate the experimental data, whereas lines represent the linear fits for $T \rightarrow 0$. The inset shows the field dependence of the slope of the low-$T$ fits ($A$ coefficient). (b) $H$-$T$ phase diagram of CeTiGe below 4.3 K deduced from $M(H)$, $\rho(H)$, and $\Delta l(H)/l_0$. The hatched area indicates the hysteresis region.

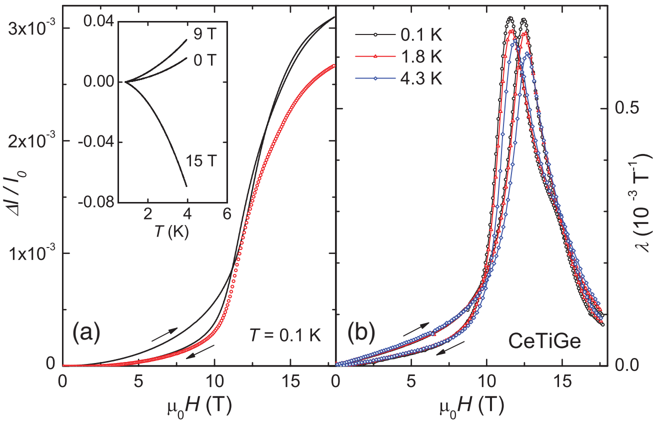

FIG. 4. (Color online) (a) Linear magnetostriction $\Delta l(H)/l_0$ measured up to $\mu_0 H = 18$ T at $T = 0.1$ K for two perpendicular sample orientations (black lines and red circles). Inset: The linear thermal expansion $\Delta l(T)/l_0$ vs $T$ is plotted for $1 < T < 4$ K and three selected fields of 0, 9, and 15 T: $\Delta l(T)/l_0$ changes sign across the MMT. (b) Plot of the magnetostriction coefficient $\lambda(H) = (1/\mu_0 l_0)\partial l/\partial H$ vs $H$ for the up and down sweeps at 0.1, 1.8, and 4.3 K.

below and above $H_c$. All curves follow the $T^2$ power law expected for a FL over two decades of temperature. All plots for fields below the MMT show almost the same slope, being just slightly shifted upward with increasing field. In contrast, the slope of the plots for $H > H_c$ is much smaller. In a FL picture this slope is related to the quasiparticle-quasiparticle scattering cross section, and it is proportional to the square of the quasiparticle effective mass. In CeTiGe this relation was shown to be fulfilled at $H = 0$.[12] Thus, the drop from a large slope for $H < H_c$ to a small slope for $H > H_c$ indicates a pronounced drop of the effective mass of the quasiparticles. For a quantitative analysis we fitted the low-$T$ part ($T < 1.5$ K) with the function $\rho(T) = \rho_0 + A(H)T^2$, and plotted in the inset of Fig. 3(a) the field dependence of the $A$ coefficient. It drops from 0.6 to 0.06 $\mu\Omega$ cm K$^{-2}$, which implies at least a decrease of the effective mass by a factor of 3. Hence, the MMT is linked to a pronounced decrease of the quasiparticle density of states at the Fermi level. A further analysis demonstrates that the magnetoresistance at fields $H < H_c$ follows the behavior expected for a normal metal with a FL ground state. At all temperatures investigated here $\rho(H)$ first increases quadratically but then evolves toward a linear $H$ dependence before $H$ approaches $H_c$. The region with quadratic $H$ dependence is larger for higher $T$. This suggests a scaling behavior. We therefore plotted the data according to Kohler's rule: $[\rho(H) - \rho(0)]/\rho(0) = f[H/\rho(0)]$ (not shown). All the data for $T = 0.02$, 0.5, and 1.8 K fall onto the same curve, almost up to $H_c$, while the data for $T = 3.2$ and 4 K exhibit only a slight upward deviation. Thus, for $H < H_c$, $\rho(H)$ follows perfectly Kohler's rule, confirming a FL state and the absence of significant spin disorder scattering.

In metallic systems metamagnetic transitions from a paramagnetic to a polarized state are usually connected with a large magnetostriction.[17] We therefore investigated the effect of the magnetic field on the length of a CeTiGe sample. The linear magnetostriction $\Delta l(H)/l_0$ was measured on a CeTiGe polycrystal with a length of 4 mm at different temperatures between 0.1 and 4.3 K. Afterward, the sample was rotated by 90° and the measurements were repeated. The relative length changes $\Delta l(H)/l_0$ vs $H$ observed at 0.1 K are plotted in Fig. 4(a). The field dependence of the magnetostriction is completely dominated by a huge, steplike increase in the field range between 10 and 15 T. In contrast, below 10 T the magnetostriction is comparatively small, while saturation seems to occur beyond 18 T. Thus, the MMT is connected with a steplike increase in $\Delta l(H)/l_0$ of the order of $2.0 \times 10^{-3}$, which is almost one order of magnitude larger than the one observed in CeRu$_2$Si$_2$.[18] Rotating the sample does not affect the overall behavior, and only the magnitude gets slightly reduced, indicating that the polycrystals do not have a significant texture. Furthermore, the curves for increasing and decreasing field differ slightly, showing a hysteresis. This is better seen in the linear magnetostriction coefficient $\lambda(H) = (1/\mu_0 l_0)\partial l/\partial H$ shown in Fig. 4(b). It displays sharp peaks at $\mu_0 H_c \approx 12.3$ and 11.4 T for increasing and decreasing fields. The hysteresis width is about 0.9 T. As in magnetization, there is almost no change with increasing temperatures up to 4.3 K, except for a small decrease of the peak height.

According to magnetoelastic theory one expects $(1/\mu_0 V)\partial V/\partial H = KM^2(H)$, where $K$ is the magnetoelastic coupling constant.[19] A preliminary analysis reveals such a quadratic dependence both at low and at high fields. $K$ has a similar value below and above the transition, of the order of $5 \times 10^{-3}(\mathrm{Ce}/\mu_\mathrm{B})^2$, which is of the same size as those reported for the 3$d$ metamagnetic systems YCo$_2$ and LuCo$_2$.[17]

We also performed preliminary measurements of the thermal expansion. In the inset of Fig. 4 we show the thermal expansion $\Delta l(T)/l_0$ at constant fields of 0, 9, and 15 T, i.e., well below, just below, and above the MMT, for $0.05 < T < 4$ K. The thermal expansion is positive at $H = 0$, becomes larger with increasing field, but changes sign and becomes strongly negative for $H > H_c$. The large positive thermal expansion at zero field is typical for Ce-based KL systems. In a simple approach this can be related to the larger volume of the trivalent state compared to that of an intermediate valent state, and to the slight shift of the valence toward 3+ with increasing temperature. A pronounced change at the MMT from a large positive thermal expansion to a large negative one has already been observed in $CeRu_2Si_2$ and is also, in a simple approach, related to the field-induced stabilization of a "localized" trivalent state. An analysis of the thermal-expansion coefficient $\alpha(T) = (1/l_0)\partial l/\partial T$ plotted as $\alpha(T)/T$ vs $T^2$ (not shown) shows the ratio $\alpha(T)/T$ to be almost independent on $T$, i.e., the thermal expansion is completely dominated by the linear-in-$T$ electronic contribution expected in a FL. The ratio $\alpha(T)/T$ increases with $H$, from $2 \times 10^{-6}$ $K^{-2}$ at $H = 0$ up to $4.6 \times 10^{-6}$ $K^{-2}$ for fields just below $H_c$, but then switches to a large negative value of about $-11 \times 10^{-6}$ $K^{-2}$ at fields just above $H_c$. Further increasing the field leads to a reduction of the absolute value, with $\alpha(T)/T = -3.3 \times 10^{-6}$ $K^{-2}$ at $\mu_0 H = 18$ T. The weak $T$ dependence of $\alpha(T)/T$ below 4 K in CeTiGe, even for fields close to the critical field, contrasts the large $T$ dependence reported for $CeRu_2Si_2$ close to $H_c$ in the same $T$ range.[20,21] This likely reflects the difference between a continuous crossover in $CeRu_2Si_2$, which implies strong $T$-dependent fluctuations near the critical field, and the first-order transition in CeTiGe, for which no fluctuations are expected.

An interesting parameter in KLs is the Grüneisen ratio $\Gamma = (V_{mol}/k)\beta(T)/C(T)$, where $k$ is the compressibility, $V_{mol}$ the molar volume, and $\beta(T)$ the volume thermal-expansion coefficient. This ratio between $\beta(T)$ and $C(T)$ reflects the pressure dependence of the characteristic energy of the system.[22] With $\beta(T)/T = 3\alpha(T)/T = 6 \times 10^{-6}$ $K^{-2}$, $\gamma = 0.3$ J/$K^2$ mol, $V_{mol} = 41 \times 10^{-6}$ $m^3$/mol, and assuming $k = 100$ GPa (a typical value for intermetallic Ce-based systems), we obtain a Grüneisen ratio $\Gamma \approx 85$ at zero field. Such a large $\Gamma$ is typical for KLs close to the transition between a magnetic and a paramagnetic ground state.[22] However, it is a factor of 2 smaller than in $CeRu_2Si_2$, likely reflecting the absence of critical behavior in CeTiGe.

In Fig. 3(b) we show the preliminary $H$-$T$ phase diagram as deduced from the magnetization, magnetoresistance, and magnetostriction results. The critical fields obtained from the peak positions in the $H$ derivative of these properties agree nicely and evidence a vertical phase boundary. Furthermore, there is no significant increase in the transition width or the hysteresis width with temperature at least up to 4.3 K, in contrast to the significant broadening observed in $CeRu_2Si_2$ in the same $T$ range. This is a further evidence for the presence of a true thermodynamic phase transition in CeTiGe.

In summary, our investigation of the paramagnetic HF system CeTiGe reveals a pronounced MMT at a moderate field $\mu_0 H_c = 12$ T. At this field we observe a large increase in the magnetization $\Delta M = 0.74\mu_B/\mathrm{Ce}$ and in the magnetostriction $\Delta l/l_0 = 2.0 \times 10^{-3}$, which are even larger than in the prototypical HF metamagnet $CeRu_2Si_2$, as well as a pronounced drop in the magnetoresistance. The opening of a hysteresis between increasing and decreasing field data in all investigated properties indicate this MMT to be a real thermodynamic transition of the first-order type, in contrast to the crossover behavior reported for $CeRu_2Si_2$. Increasing the temperature from 0.02 to 4.3 K leaves the transition almost unaffected: $H_c$ is nearly independent of $T$ and neither the hysteresis width nor the transition width increases with $T$, which is incompatible with crossover behavior. In addition, the thermal expansion in fields close to $H_c$ do not show evidence of critical fluctuations up to 4.2 K, giving further support to the first-order type of the transition. The analysis of the resistivity and magnetoresistance data points to a pronounced drop of the effective mass of the electronic quasiparticles across the transition, while thermal expansion and magnetostriction suggest that this drop might be connected with some kind of localization of the 4*f* electrons. The large Grüneisen parameter $\Gamma \approx 85$, deduced from the thermal-expansion and specific-heat data, is in accordance with CeTiGe being close to the transition from a paramagnetic to a magnetically ordered ground state. The first-order nature of the MMT as well as the exceptionally large anomalies in $M(H)$ and $l(H)$ establish CeTiGe as a rather unique metamagnetic system among the 4*f*-based Kondo-lattice compounds.

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