Type-II superconductivity in a dilute magnetic system: $La_{1-x}Tm_xRu_3Si_2$

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Superconducting and magnetic properties of $La_{1-x}Tm_xRu_3Si_2$ ($x=0$, 0.08, and 0.16) have been investigated through measurements of both electric and magnetic properties. Tm is in a trivalent state and carries a magnetic moment $\approx 8\mu_B$. We find that the magnetic measurements are not suited to the determination of the critical line $H_c(T)$ for the onset of type-II superconductivity in materials where one has at the same time superconducting diamagnetic shielding and paramagnetic contribution due to the localized spins. On the other hand, transport experiments are much less ambiguous, and allow us to determine accurately the critical temperature in a given applied magnetic field. In particular, they allow us to suggest a more appropriate interpretation of the maximum in the magnetic-susceptibility curve already observed in materials of the same family: this maximum results from a competition between the incomplete diamagnetic shielding and the paramagnetic contribution from the magnetic ions in the mixed phase, rather than from a spin-glass freezing of the paramagnetic ions. In zero field, the rate of depression of the critical temperature upon substituting nonmagnetic La ions by magnetic Tm ions is $dT/dx=-8\pm1$ K/at. % Tm.

I. INTRODUCTION

The series RRu_3Si_2 (R stands for rare earth) is an interesting family of ternary compounds that includes various superconductors, for $R=$La,Y,Th,Ce, with their transition temperature T_c ranging from 7.3 K for $R=$La,[1] to 1 K for $R=$Ce.[2] On the other hand, the compounds with $R=$Tm,Nd are magnetic, but not superconducting. The hexagonal crystallographic structure of these materials[3] is illustrated in Fig. 1. Ru atoms are located in two-dimensional sheets of triangular clusters. The Ru-Ru distance within a cluster is smaller than in the bulk ruthenium metal. An important feature is the apparent doubling of the unit cell along the c direction. This is due to a small distortion of the Ru clusters, which the structure repeats with a periodicity $c=2c_0$, with c_0 the lattice parameter if Ru clusters were undistorted. This feature differentiates this structure from the $CeCo_3B_2$ structure where such a distortion of the Co clusters does not exist. These structural properties may be responsible for T_c being unusually high in $LaRu_3Si_2$. As the members of this series which have magnetic rare-earth (MR) atoms are not superconducting, the solid solutions $La_{1-x}MR_xRu_3Si_2$ form an interesting system suitable for the study of the competition between magnetism and superconductivity. $La_{1-x}Ce_xRu_3Si_2$ in the full range $0\le x\le 1$ (Ref. 4) is a system apart, because Ce is nonmagnetic in this material. The other system which has been studied is $La_{0.95}Gd_{0.05}Ru_3Si_2$,[5] where $MR=$Gd is indeed magnetic, since trivalent Gadolinium carries a spin $\frac{7}{2}$. Coexistence of superconductivity and spin-glass freezing was inferred from a preliminary study of mag-

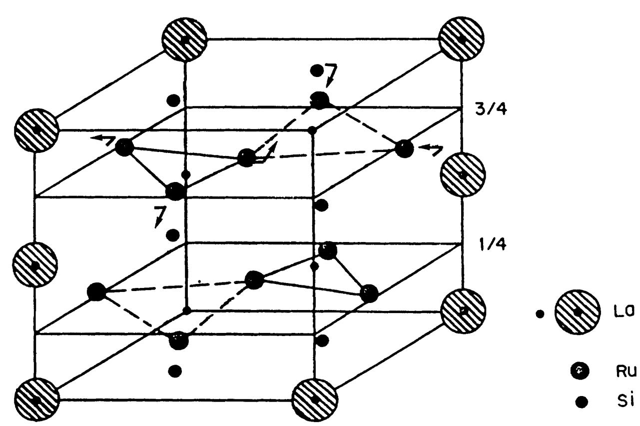

FIG. 1. Hexagonal structure of $LaRu_3Si_2$ ($a=5.67$ Å, $c=7.120$ Å). The structure is characterized by two-dimensional sheets of triangular clusters of Ru atoms and linear chains of La atoms (after Ref. 3).

netic properties of this system.

In the present work, we investigate another solid solution with $MR = \mathrm{Tm}$, for $x = 0$, 0.08, and 0.16. Magnetic and transport properties have been measured for applied magnetic fields H up to 12 kG. All the samples are type-II superconductors, and differ only by their characteristic parameters, namely flux exclusion, superconductivity transition temperature $T_c(H)$, and higher critical field $H_{c2}(T)$. Our studies suggest that the maximum in the magnetic susceptibility curve, attributed to a spin-glass freezing in the system $La_{0.95}Gd_{0.05}Ru_3Si_2$,[5] is more likely attributable to the progressive onset of type-II superconductivity.

Both magnetic and transport properties have been investigated to analyze the superconducting behavior. We first report results of these experiments for the $LaRu_3Si_2$ matrix in Sec. III. Modifications of these properties upon introducing Tm ions in the matrix are reported in Sec. IV.

II. EXPERIMENT

$La_{1-x}Tm_xRu_3Si_2$ ($x = 0, 0.08, 0.16$) samples have been prepared by melting the elements in an arc furnace under argon atmosphere. The nominal composition of each alloy was $La_{1-x}Tm_xRu_{3.5}Si_2$, the excess of ruthenium preventing the formation of the $LaRu_2Si_2$ phase.[1] Loss of weight during the melting was less than 0.5%. Lattice parameters deduced from the x-ray-diffraction pattern decrease with x, ranging from $a = 5.675$ Å, $c = 7.115$ Å for $x = 0$, to $a = 5.635$ Å, $c = 7.080$ Å for $x = 0.16$. Homogeneity of the samples has been checked by microprobe analysis. Magnetic susceptibility curves in fields $H < 2000$ G have been measured with a Faraday balance. The other magnetic properties have been measured on a MANICS magnetometer-susceptometer model DSM8. In-field electric resistivity has been measured by a capacitor bridge method described elsewhere.[6] Basically, the sample and a reference resistance are fed with an alternate current source at low frequency (150 Hz). After a suitable amplification, the signals at the pins of the samples and of the reference resistance are 180° phase shifted and then mixed. The output of the mixer is connected to a synchron amplifier acting as a null detector. The method allows a measurement of resistances $\approx 10^{-3}$ Ω within 10^{-5} Ω.

III. $LaRu_3Si_2$

In the normal phase, magnetic susceptibility $\chi(T)$ of $LaRu_3Si_2$ comes from the Pauli contribution of the conduction electrons, and is almost temperature independent. Deviation from the Pauli behavior can be observed upon cooling; $\chi(T)$ drops when the system enters in the superconducting state. This is illustrated in Fig. 2, where we have reported several $\chi(T)$ curves, obtained by cooling the sample under constant magnetic field H, for different values of this parameter. Throughout this paper, we define $\chi = M/H$, with M the magnetization. It should not be confused with the slope dM/dH of the magnetization curves at low temperatures where the magnetization is not a linear function of H in the range of magnetic fields investigated. Let χ_n designate the susceptibility in the normal state (i.e., in the nonsuperconducting state). We define the superconducting transition temperature T_c, as the temperature where deviation of $\chi(T)$ from $\chi_n(T)$ takes place. For each value of H, we then determine the transition temperature, marked by an arrow in Fig. 2, which defines the coordinates (H,T) of a point of the critical line in the phase diagram. Along this line shown in Fig. 3, H is the field at which the system undergoes the superconducting-normal phase transition at temperature T. It is then the upper critical line $H_{c2}(T)$ of the type-II superconductor.

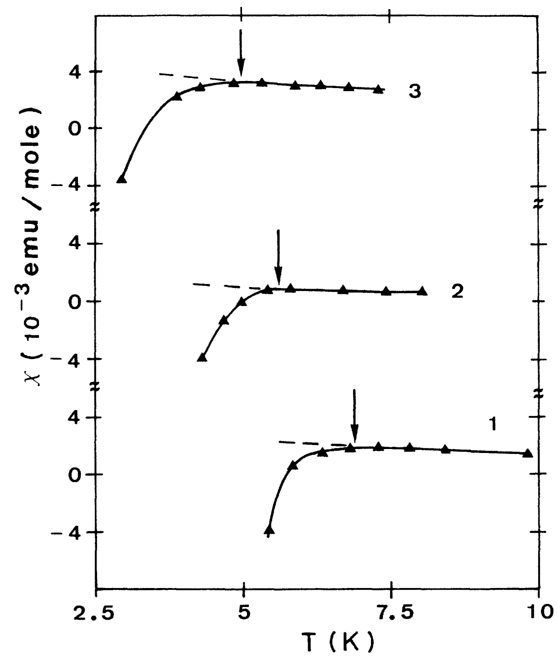

FIG. 2. Magnetic susceptibility curves of $LaRu_3Si_2$ at magnetic fields $H = 2237$ G, 8600 G, and 10 785 G, for curves 1 to 3, respectively. Arrows point the temperature of the normal-superconducting transition, determined by the departure of the susceptibility from its normal behavior (broken line).

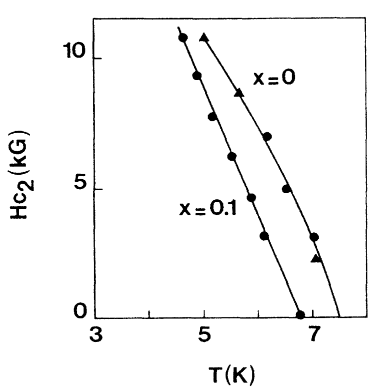

FIG. 3. Upper critical field H_{c2} as a function of temperature in type-II superconductors $La_{1-x}Tm_xRu_3Si_2$ ($x = 0$ and 0.08), as deduced from resistivity (full triangles) and magnetic susceptibility (full dots).

Magnetic measurements have been complemented with the investigation of transport properties. Some resistivity curves $\rho(T)$ are reported in Fig. 4 for several values of the static magnetic field applied to the sample. The resistivity in the normal state is nearly independent of T in the range $9<T<20$ K, because at such low temperatures, ρ reduces to ρ_n, the residual resistivity of the metal in its normal state. ρ_n is also independent of H up to the highest field available in the experiments (12 kG), as is expected for a nonmagnetic material. At lower temperatures, however, ρ decreases and vanishes in the superconducting state. Since the variation of ρ between $\rho \approx 0$ and $\rho \approx \rho_n$ is spread out over a temperature range of the order of 2 K, we face the question of how to determine T_c from the resistivity curves. In such circumstances, T_c is commonly defined as the temperature where ρ is a given fraction of ρ_n. The choice of this ratio, however, is arbitrary, ranging from $\rho/\rho_n \approx \frac{1}{2}$ [which would roughly correspond to the inflection point of the $\rho(T)$ curves] to $\rho/\rho_n \approx 1$ [which would correspond to the onset of deviations from the $\rho_n(T)$ behavior]. In the present case, however, it can be determined unambiguously from the comparison between magnetic and transport properties: we choose it so that the corresponding temperature matches the transition temperature deduced from the $\chi(T)$ curves in Fig. 2 at the same magnetic field. The result is $\rho/\rho_n=0.77$, hence the equation of the $H_{c2}(T)$ critical line:

$$\rho(H_{c2},T)=0.77\rho_n \ . \tag{1}$$

The points of the $H_{c2}(T)$ line in the phase diagram deduced from the resistivity curves, according to Eq. (1), are reported in Fig. 3, which complements the data available from the $\chi(T)$ curves. We find $T_c=7.5$ K for $H_{c2}=0$, in reasonable agreement with the value 7.3 K reported earlier in this paper.[1] Note that at $T \to T_c$, the slope of the $H_{c2}(T)$ line is finite, as is seen in Fig. 3, i.e.,

$$H_{c2} \propto (T-T_c) \ . \tag{2}$$

This is indeed expected for a type-II superconductor, as H_{c2} is related to the coherence length ξ according to[7]

$$H_{c2}=\Phi_0/[2\pi\xi^2] \tag{3}$$

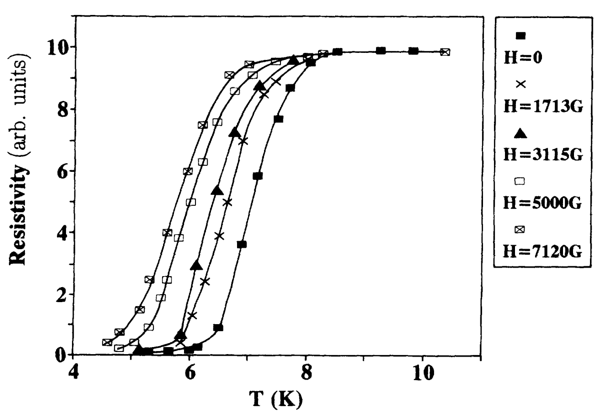

FIG. 4. Resistivity curves for $LaRu_3Si_2$ in the vicinity of the critical temperature for various magnetic fields H applied to the sample.

with $\Phi_0=\hbar/(2e)$ the flux quantum. Within the Ginzburg-Landau theory, the critical exponents are classical, i.e., $\xi(T) \propto (T-T_c)^{-1/2}$. Thus Eq. (2) follows from Eq. (3). The linear temperature dependence of H_{c2} is expected only in the vicinity of T_c. Its extrapolation down to $T=0$, as it is done sometimes (see, for example, Ref. 8), leads to an overestimation of $H_{c2}(0)$ since $H_{c2}(T)$ should saturate at low temperatures. Therefore, from Fig. 3, only the order of magnitude $H_{c2}(0) \approx 2 \times 10^4$ G can be estimated.

The lower critical field H_{c1} at which the first flux enters the superconductor is related to H_{c2} according to the relation[9]

$$\frac{H_{c1}}{H_{c2}}=\frac{\ln\kappa}{2\kappa^2} \tag{4}$$

with $\kappa=\lambda/\xi$ the Landau-Ginzburg parameter, λ being the penetration depth of the magnetic field. From the experimental point of view, H_{c1} can be defined as the field at which the magnetization curve $M(H)$ departs from the linear behavior. We find $H_{c1}(0) \approx 10^2$ G; hence, after Eq. (4), $\kappa \approx 10$. This material is thus a typical type-II superconductor with a large value of κ. H_{c1} is too small to be measured close to T_c with our experimental setup. Nevertheless, the linear temperature dependence of H_{c2} observed in the present work implies a similar behavior of $H_{c1}(T)$ after Eq. (4), if we assume that κ is temperature independent. Such a linear behavior for $H_{c1}(T)$ has been observed in other type-II superconductors with large κ.[8] We then confirm previous claims[8] that the conventional Ginzburg-Landau theory predicting linear temperature dependences of the critical fields is consistent with a local limit BCS form in type-II superconductors.

IV. $La_{1-x}Tm_xRu_3Si_2$

A. Magnetic properties

At high temperatures, magnetic susceptibility curves $\chi(T)$ of the $x=0.08$ and $x=0.16$ samples exhibit a Curie-Weiss behavior, with paramagnetic Curie temperatures Θ_p 13.7 K and 18.4 K, respectively. Effective magnetic moment of Tm, deduced from the Curie constant, is $8.6\mu_B$ and $8.1\mu_B$ for $x=0.08$ and $x=0.16$ samples, respectively. This moment is larger than the theoretical value ($7.5\mu_B$) predicted by Hund's rule for the Tm^{3+} ion, and suggests that there is a significant conduction electron spin polarization around Tm^{3+} magnetic ions. Our L_3-edge measurements on $TmRu_3Si_2$ show only one white line at the threshold energy of the Tm^{3+} ground configuration, and we conclude that Tm is nearly trivalent in $La_{1-x}Tm_xRu_3Si_2$ in these systems. However, the 10% increase of the magnetic moment in the $x=0.08$, 0.16 samples with respect to the Tm^{3+} free-ion value gives evidence for a non-negligible hybridization of the $4f$ states with the conduction electron states of the matrix.

At low temperatures, $\chi(T)$ curves show a quite different behavior, as illustrated in Figs. 5 and 6 for the $x=0.08$ and $x=0.16$ samples with the static magnetic

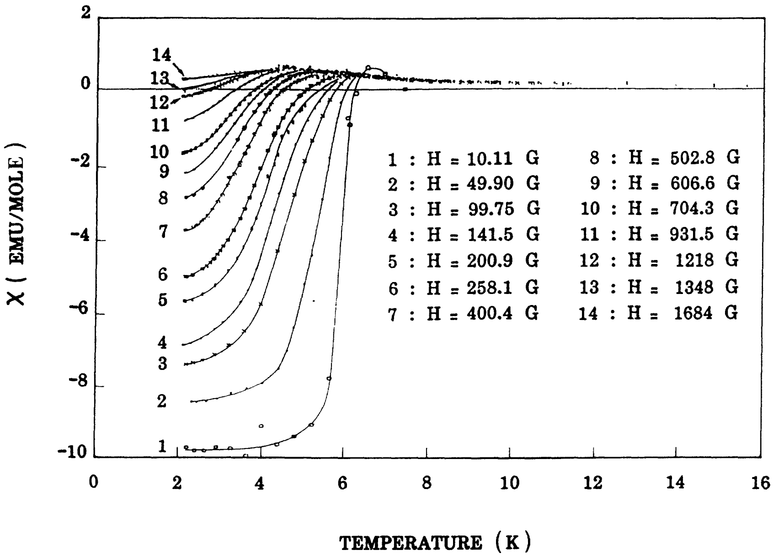

FIG. 5. Zero-field-cooled magnetic susceptibility of $La_{0.92}Tm_{0.08}Ru_3Si_2$ as a function of temperature for several magnetic fields in the range 0–1700 G.

field H as a parameter. Note the experimental conditions must be specified in the mixed phase, i.e., in the region between $H_{c1}(T)$ and $H_{c2}(T)$ lines of the phase diagram, since magnetic irreversibilities due to vortex motion are important. The $\chi(T)$ curves in Figs. 5 and 6 are zero-field cooled curves, i.e., the sample is first cooled from a temperature significantly higher than T_c down to 2 K in zero magnetic field and zero-field gradient. Then the magnetic field H is applied to the sample, and $\chi(T)$ is measured upon warming the sample at a typical rate 2 K per hour. In the normal state, $\chi(T)$ increases upon cooling according to the Curie-Weiss contribution from the Tm localized spins. In the superconducting phase $\chi(T)$ decreases while decreasing the temperature, and changes sign, due to the diamagnetic shielding. As a result, $\chi(T)$ goes through a maximum at a temperature which we call T_m. This temperature decreases as H increases.

The shape of the $\chi(T)$ curve near T_m depends on H.

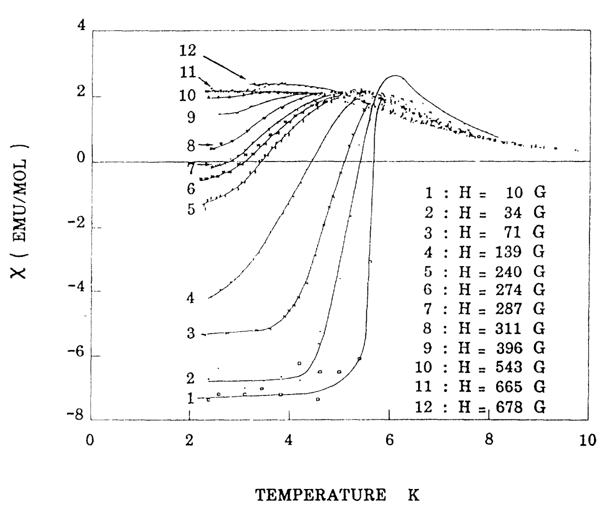

FIG. 6. Zero-field-cooled magnetic susceptibility of $La_{0.84}Tm_{0.16}Ru_3Si_2$ as a function of temperature for several magnetic fields in the range 0–678 G.

At low applied fields, one observes a peak which turns into a rounded maximum at relatively higher fields. We now comment on the origin for such an effect of H. In the normal state at low temperatures, onset of local magnetic correlations results in magnetic clustering effects yielding a Langevin-type contribution to the magnetization. As the effective magnetic moment associated with these magnetic clusters is large, this part of the magnetization is easily saturated in the magnetic field for $T<10$ K. This saturation means a negative curvature of the magnetization curve $M(H)$, and thus a decrease of χ_n as a function of H, at a given temperature, as it is observed and shown in Figs. 5 and 6. This decrease of χ_n, combined with the decrease of T_m, explains the rounding of the high-temperature wing of the peak in $\chi(T)$ as H increases. The rounding of the low-temperature wing has a different origin. The upper critical line $H_{c2}(T)$ and the lower critical line $H_{c1}(T)$ intersect at $(H=0, T=T_c)$ for a type-II superconductor which, like our sample, does not belong to the family of high-T_c superconductors. In the limit $H \rightarrow 0$, the magnetic susceptibility will thus shift abruptly from its positive value in the normal state to the negative value, characteristic of the Meissner effect upon cooling through T_c. This explains the very sharp variation of $\chi(T)$ in the vicinity of T_c at the lowest magnetic field investigated in Figs. 5 and 6. As H increases, however, there is a finite range of temperatures where the mixed phase is stable, in which χ varies continuously as a function of T, hence the rounding of the low-temperature wing of the peak in $\chi(T)$ when H increases.

A more detailed analysis of the magnetic properties can be achieved from the investigation of magnetization curve $M(H)$. Such a curve is illustrated in Fig. 7 for the $x=0.08$ sample, at low temperature. M is negative and decreases algebraically as H increases up to some value H^+ as expected, but then M increases with H. This is evidence of a significant paramagnetic contribution arising from a spin polarization of the Tm impurities. Indeed in the normal state, magnetic impurities polarize the electron gas in their vicinity, due to the magnetic ex-

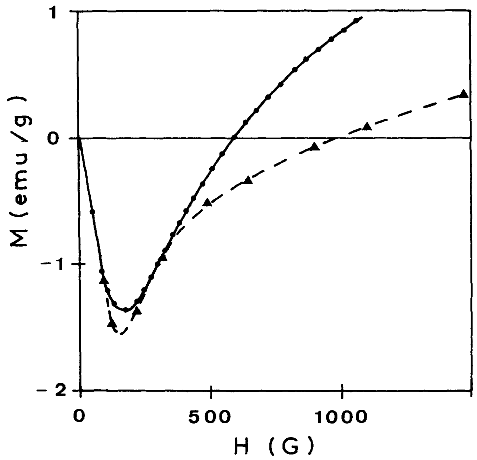

FIG. 7. Virgin magnetization curve of $La_{0.92}Tm_{0.08}Ru_3Si_2$ in the superconducting phase at temperature $T=3.2$ K (broken curve, triangles) and 4 K (full curve, dots).

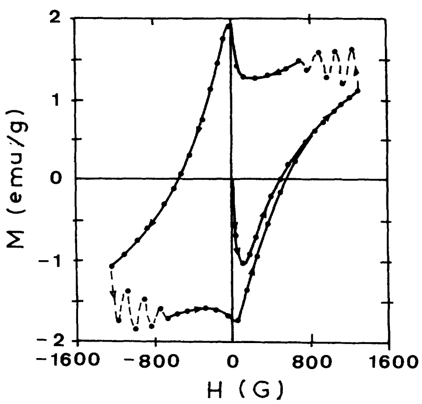

FIG. 8. Hysteresis cycle of $La_{0.92}Tm_{0.08}Ru_3Si_3$ at temperature $T=4.3$ K. The cycle starts at $M=0$, $H=0$ (zero-field-cooled state) then the arrows indicate how the cycle has been obtained. The broken part corresponds to the region where oscillations are detected (see text).

change between the localized spin of the impurity and the spins of itinerant electrons. We have already noticed that the large value of the magnetic moment carried by the Tm ions gives evidence of the ferromagnetic cloud of the electron gas spin polarization surrounding the magnetic ions. Since magnetism opposes superconductivity, the pinning of Abrikosov vortices on the magnetic impurities realizes the stable configuration, as the core region of a vortex is in a quasinormal state. Since the amplitude of the Friedel oscillations of electron spin polarization in a normal metal scales like r^{-3} as a function of the distance from the localized spin, the effective interaction between a vortex and a magnetic impurity is long range, and attractive. Note also that the core region of a vortex is also the region where the magnetic field penetrates, so the pinning of a vortex on a magnetic impurity also realizes the configuration which minimizes the magnetic energy associated to the Zeeman term of the Hamiltonian. On one hand this effect contributes to the pinning of a vortex on a magnetic impurity. On another hand the magnetic impurity will be spin polarized by the magnetic field which penetrates the core region of the vortex in the mixed phase. We can then understand the magnetization curves $M(H)$ at given temperature $T<T_c$ as follows. At low magnetic fields, M is negative and decreases in algebraic value as a result of the diamagnetic shielding, and goes through a minimum at $H=H^+$. At larger fields, M increases with H, due to the paramagnetic contribution of the magnetic ions in the core region of the vortices in the mixed phase, and does not saturate for two reasons. First, the applied magnetic field is not large enough so that the spin polarization of the localized spins in the core region of the vortices is not saturated. Second, as H increases, so does the concentration of vortices in the mixed phase and therefore the fraction of localized spins involved in the core regions penetrated by the magnetic field. Only at still larger magnetic fields will the upper critical field $H_{c2}(T)$ be reached, above which the magnetization curve will match that of the normal state.

Figure 8 illustrates the magnetic hysteresis curve of the $x=0.08$ sample at 4.3 K. Jumps of magnetization and oscillations are observed (broken curve in Fig. 8). They are characteristic of type-II superconductivity, and take their origin in the motion of vortices induced by a variation of the magnetic field in the mixed phase. This motion implies a dissipation, and thus a local heating, which in turn favors the penetration of vortices in this region, a source of additional heating. Instability can take place when this heating is larger than the initial fluctuation of temperature. These magnetothermal instabilities can be evidenced either as jumps in the magnetization or as jumps of temperature, as the magnetic field is varied. They have been first evidenced in the 1960s,[10–12] due to their importance in the technology of superconducting coils. Similar effects have also been observed in high-T_c superconductors,[13] and recent systematic studies have elucidated their origin.[14] The study of such instabilities is beyond the scope of the present work, and would require a record of the magnetization data at interval steps which are small with respect to the period of the oscillations, in contrast with the data in Fig. 8.

B. Transport properties

For the $x=0.08$ and $x=0.16$ samples, the transition between superconducting and normal state occurs in the range of temperatures $T<\Theta$ where the Curie-Weiss law does not apply, so that the law for $\chi_n(T)$ is basically unknown. As a consequence, it becomes impossible to determine when the experimental curve $\chi(T)$ deviates from $\chi_n(T)$. This is a major difference with the $x=0$ sample, in which the onset of such deviation was used to determine T_c. Since $\chi_n(T)$ increases upon cooling, T_m is a lower limit for the superconducting transition temperature. Still in the low-field limit, the peak in $\chi(T)$ is sharp, so that T_m becomes a reasonable estimate of the transition temperature. As H increases, however, we have argued that the peak turns in a rounded maximum, in which case the transition temperature is much larger than T_m and cannot be determined from magnetic measurements. Transport experiments then become of primary importance to determine T_c.

The resistivity curves are illustrated in Fig. 9 for $x=0.08$, as a function of T, for different values of H. The curves have the same S shape as in the $x=0$ sample. In the normal state, above 7.5 K, $\rho(T)$ saturates at a value ρ_n which depends on H only slightly, due to a small positive magnetoresistance. Also ρ_n increases with x, as Tm ions act as impurities which contribute to the residual resistivity of the metal, in the normal state. This is best evidenced in Fig. 10 where we have reported $\rho(T)$ at $H=0$ for both $x=0.08$ and $x=0.16$ samples, for comparison. We then assume that Eq. (1) derived for $x=0$ still holds true at $x\neq 0$, and use it to determine the transition temperature at any magnetic field H. The results are displayed in Fig. 4 for $x=0.08$. T_c is depressed by 0.7 K and 1.4 K for $x=0.08$ and $x=0.16$, respectively, with respect to $x=0$. Taking into account the uncertainty in the determination of x from microprobe analyses, we then find $dT_c/dx=-8\pm 1$ K per atomic fraction of Tm.

It is clear from our studies that the transport studies are essential to determine unambiguously the superconducting transition temperature $T_c(H)$ in the materials of the kind that are of interest here. For example, for the sample $x=0.08$, χ is positive down to the lowest temperature investigated for $H>1600$ G (see curve 14 at $H=1684$ G in Fig. 5). The magnetic susceptibility, as we mentioned above, consists of two opposing contributions, a paramagnetic contribution coming from the magnetic impurities, and the diamagnetic contribution due to superconductivity. At this field, the net susceptibility remains positive at all temperatures. Thus, it is difficult to infer $T_c(H)$ from the susceptibility measurements. On the other hand, we infer from the in-field resistivity curves (Fig. 9) that $T_c(H=1684\ \text{G})\approx 6$ K. If we call $T_0(H)$ the temperature at which χ changes sign, then it is clear that $T_c(H)>T_m(H)$ and also $T_c(H)>T_0(H)$. In

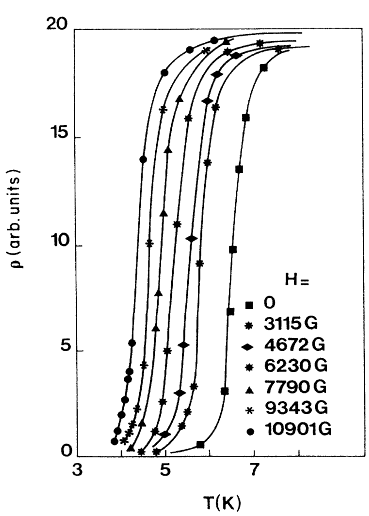

FIG. 9. Resistivity curves of $La_{0.92}Tm_{0.08}Ru_3Si_2$ for different values of the applied magnetic field H.

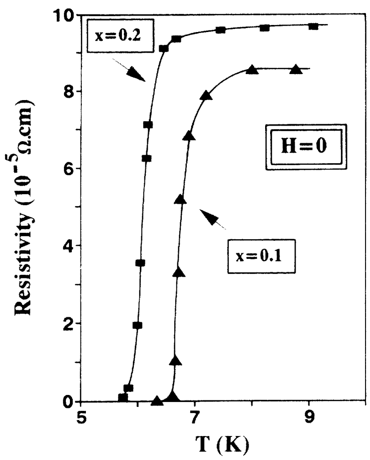

FIG. 10. Resistivity curves of $La_{1-x}Tm_xRu_3Si_2$ at zero magnetic field, for $x=0.08$ and 0.16.

particular, we find the upper critical field $H_{c2}(T\to 0)$, inferred from our measurements for the sample $x=0.08$, is larger than the highest field (11 kG) available in our experiments; this is one order of magnitude larger than the value resulting from the assumption $T_c(H)=T_0(H)$.[5] We therefore find that the transport experiments are best suited to determine the superconducting transition temperature in the presence of magnetic impurities.

V. DISCUSSION

If the density of electron states is not drastically modified by the introduction of the magnetic impurities in the matrix, dT_c/dx should satisfy the scaling law[15]:

$$dT_c/dx \propto J_{sf}^2(g-1)^2J(J+1)\ . \tag{5}$$

J_{sf} is the exchange coupling between the localized spin and the spin of the conduction electron. J is the angular momentum and g the Landé factor of the magnetic impurity. If we assume that J_{sf} is the same, whether the magnetic impurity is Gd or Tm, the scaling law in Eq. (5) predicts $dT_c/dx=-1$ K per atomic fraction of Tm impurities, when taking into account that $dT_c/dx=-12$ K per unit fraction of Gd impurities.[5] This is much smaller than the experimental value $dT_c/dx=-8\pm 1$ K found in this work in $La_{1-x}Tm_xRu_3Si_2$. The same procedure applied to $La_{1-x}Ce_xRu_3Si_2$ also predicts a value of $|dT_c/dx|$ much smaller than the experimental value:[4] $dT_c/dx=-6$ K. Indeed, $|dT_c/dx|$ is often anomalously large for Ce impurities and sometimes for Sm, Eu, Tm, and Yb impurities in different superconductors. This feature is attributable either to a Kondo effect,[16] or mixed valence behavior which these ions often exhibit in different matrices. Although we made it clear that Tm is trivalent in our case, the fact that the effective moment is about 10% larger than the theoretical value predicted for trivalent Tm ion suggests a significant hybridization of the conduction electrons with $4f$ electron states. The large decrease of T_c as a function of x in $La_{1-x}Tm_xRu_3Si_2$ is also consistent with the fact that the material is no longer a superconductor in the limit $x=1$, as the extrapolation of the linear variation of T_c upon x leads to $T_c=0$ for $x=0.9$. Indeed, $TmRu_3Si_2$ is an antiferromagnet, with a spin ordering temperature $T_N=7$ K.[1]

$\chi(T)$ curves reported earlier[5] on $La_{1-x}Gd_xRu_3Si_2$ are quite similar to those we have reported in Figs. 5 and 6 for $La_{1-x}Tm_xRu_3Si_2$. The maximum in $\chi(T)$, in absence of the resistivity data, was attributed to a spin-glass transition, occurring at temperature $T_g \approx T_m$. This interpretation should be modified for several reasons. First, considering that the spin ordering temperature is 7 K for $La_{1-x}Tm_xRu_3Si_2$ in the pure $x=1$ case, it is quite difficult to believe that spin ordering may occur at $T_m \approx 5$ K in a system which is tenfold diluted. Second, T_m is about the same ($\approx 5\pm 0.5$ K) for both $x=0.08$ and $x=0.16$ samples (a variation of x by a factor 2). Then a relation $T_g \approx T_m$ violates the scaling law $T_g \propto x$, which holds for the spin-glass freezing in metallic spin glasses.[17] Finally, an analysis based on many systems shows that[18] $(dTg/dx=0.04\,dT_c/dx$ according to Abrikosov-Gorkov and Ruderman-Kittel-Kasuya-Yosida interactions. In the present case, $T_g(x=0.08)$ would be the order of 30 mK. We are thus led to the conclusion that the maximum in $\chi(T)$ results from the gradual onset of superconductivity rather than spin ordering. Indeed, we have argued that the shape of the susceptibility peak, and its roundening as H increases is consistent with this interpretation.

VI. CONCLUSION

We have presented here the results of our dc-susceptibility measurements on the superconducting materials $La_{1-x}Tm_xRu_{3.5}Si_2$ ($x=0.08$, 0.16). Our L_3-edge measurements show that the Tm ions are essentially in the trivalent state. However, the effective moment of the Tm ions is $\approx 8\mu_B$, which is higher by about 10% than the free-ion value of Tm^{3+} ions. This implies a significant ferromagnetic contribution from the conduction electron spin polarization around the Tm magnetic ions. We have argued that in such systems, where the magnetic susceptibility has two contributions (one a paramagnetic contribution coming from the magnetic impurities, and the other being the diamagnetic shielding from the superconductivity), resistivity measurements provide an unambiguous determination of the superconducting transition temperature $T_c(H)$. In high applied magnetic fields, one may not see a diamagnetic susceptibility, but the resistivity does show a signature of superconductivity.

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