First-order nature of the ferromagnetism in $CeIn_2$ investigated using muon spin rotation and by systematic substitution of La for Ce

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The nature of the first-order ferromagnetic transition in binary $CeIn_2$ alloy is investigated by muon spin rotation (μSR) measurements and chemical substitution of Ce by La in the $La_{1-x}Ce_xIn_2$ ($0.9 \leqslant x \leqslant 1.0$) series of alloys. Below 22 K, the analysis of μSR spectra shows two spin precession frequencies associated with the local field at the muon site created by the surrounding ferromagnetic ordered magnetic moments. These frequencies abruptly disappear above T_C, indicating the first-order character of this transition, as previously reported. For temperatures between 22 and 24 K, the shape of the μSR spectra indicates the existence of an additional magnetic phase with features of an incommensurate magnetic structure. The presence of this magnetic phase is supported by dc(ac)-magnetic susceptibility and specific-heat results obtained on chemical diluted samples, which also show a magnetic contribution above the ferromagnetic transition. The combined analysis of these results clarifies the first-order character of the ferromagnetic transition in $CeIn_2$, based on the existence of an intermediate magnetic phase between the paramagnetic and ferromagnetic states.

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I. INTRODUCTION

First-order phase transitions are a relevant topic in condensed-matter physics and materials science from both experimental and theoretical points of view because they are related to interesting phenomena such as colossal magnetoresistance,[1] giant magnetocaloric effects,[2] and memory shape effects.[3] The order of a phase transition is also an important issue related to the understanding of quantum phase phenomena.[4,5] A typical example is those materials in which ferromagnetic quantum phase transitions display a behavior that can be well explained in terms of a first-order transition. On the theoretical side, several mechanisms have been identified that preempt ferromagnetic quantum criticality by a first-order transition.

The effect of quenched disorder on a first-order phase transition has also been a subject of considerable scientific interest since the late 1970's. There are several distinct examples: disordered-ferroelectric transitions,[6] the vortex-matter phases of high-temperature superconductors (HTSs),[7] and electronic phase separation in manganites showing colossal magnetoresistance.[8]

Very recently, we have studied the binary $CeIn_2$ alloy, which was found to be ferromagnetic below 22 K, one of the highest ferromagnetic ordering temperatures found on this kind of compound.[9] In addition, the analysis of the magnetic, thermal, and transport properties provided evidence for the first-order character of this magnetic transition on this alloy. In order to gain further insight into the first-order nature of the magnetic transition of this material in the present work, muon spin rotation (μSR) experiments at different temperatures have been performed on the binary $CeIn_2$ alloy, and the subsequent results have been further supported by measurements of the magnetic and thermal properties on the $La_{1-x}Ce_xIn_2$ ($0.9 \leqslant x \leqslant 1.0$) series of alloys. The substitution of Ce by La will result in a change of the unit-cell volume and, as is well known, this affects the interaction between the conduction electrons and the $4f$ moments, resembling the application of external pressure.[10]

II. EXPERIMENTAL DETAILS

The $La_{1-x}Ce_xIn_2$ ($0.9 \leqslant x \leqslant 1.0$) samples were prepared by arc melting suitable amounts of the starting materials La (3*N*), Ce (3*N*), In (5*N*) (all Johnson Matthey) in an arc furnace under Ar atmosphere. The analysis of the obtained x-ray diffraction patterns indicates that all the prepared alloys crystallize in the orthorhombic $CeCu_2$ type of structure, showing a linear increase of the unit-cell volume with the decrease of the Ce concentration (x), according to Vegard's law. Thermal, transport, and magnetic properties were collected at a Quantum Design physical properties measurement system (PPMS) device, in the temperature range 2–300 K and magnetic fields up to 90 kOe. The μSR experiments were performed at the Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institute, Switzerland, using the GPS spectrometer. μSR spectra were recorded using the zero-field (ZF) configuration, in the temperature range 2–100 K.

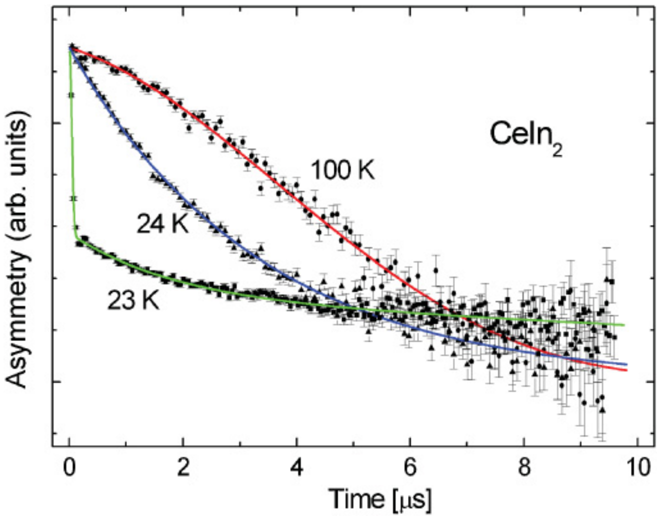

FIG. 1. (Color online) μSR spectra recorded on $CeIn_2$ at different temperatures above the ferromagnetic transition (23, 24, and 100 K).

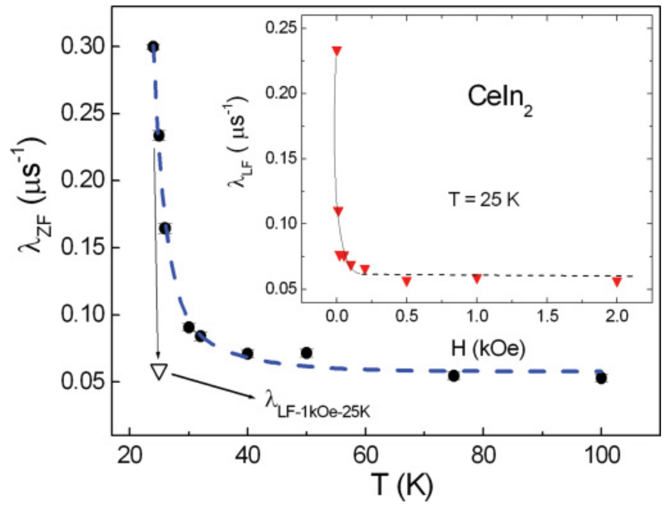

FIG. 2. (Color online) Temperature dependence of λ_{ZF} for $CeIn_2$ without applied magnetic field. For comparative purposes, λ_{LF} at 25 K and 1 kOe is also indicated. The lines are just guides to the eye. The inset details the field dependence of λ_{LF} at 25 K.

III. RESULTS

A. μSR measurements in the $CeIn_2$ alloy

The μSR spectroscopy has been extensively employed to investigate the nature of the ground state of many magnetic systems, in particular Ce, Yb, and U compounds.[11] Very recently, this technique successfully proved the first-order ferromagnetic transition of the itinerant helimagnet MnSi.[12] Due to its microscopic character and sensitivity to extremely small internal fields, it can reveal spatially inhomogeneous magnetic features associated with the influence of the crystalline environment on the muon spin motion. For this reason, we have used this useful tool in the investigation of the nature of the first-order ferromagnetic transition in the $CeIn_2$ alloy.

Typical μSR spectra recorded at different temperatures, and in a zero applied magnetic field, are depicted in Fig. 1. At temperatures well above the transition temperature ($T > 24$ K), the shape of the μSR spectra is almost Gaussian, indicating that the depolarization of the implanted muons is due mainly to the static (in the time window of the μSR experiment) field distribution created by the nuclear magnetic moments of the In atoms. Only a small contribution to the depolarization arises from the fast-fluctuating electronic magnetic moments of the Ce ions. It is interesting to note that, for $T = 23$ K, a drastic change in the spectra is observed. Its origin, as well as the implications on the magnetic behavior of the $CeIn_2$ alloy, will be discussed below. On the other hand, the μSR spectra recorded in a zero applied field above 24 K can be well fitted by

$$AP(t) = A_{\text{para}}\left[\tfrac{1}{3} + \tfrac{2}{3}(1-\sigma^2 t^2)e^{-\frac{\sigma^2 t^2}{2}}\right]e^{-\lambda_{\text{ZF}} t}, \qquad (1)$$

where A_{para} is the so-called asymmetry, which indicates the amplitude of the μSR signal. The parameter $P(t)$ has been chosen as a product of a so-called Kubo-Toyabe (KT) depolarization function and an exponential term. The KT term, $\frac{1}{3} + \frac{2}{3}(1-\sigma^2 t^2)e^{-\frac{\sigma^2 t^2}{2}}$, accounts for the depolarization of the muon ensemble by the isotropic Gaussian field distribution of width $\frac{\sigma}{\gamma_\mu}$ created by the In nuclear-magnetic moments, and γ_μ is the gyromagnetic ratio of the muon. The exponential term $e^{-\lambda_{\text{ZF}} t}$ describes the second channel of depolarization, due to the fast-fluctuating electronic magnetic moments. In the limit of uncorrelated fast fluctuations, λ_{ZF} is inversely proportional to the fluctuation rate of the electronic magnetic moments and is field independent. The parameter λ_{ZF} increases slightly upon decreasing temperature down to about 30 K, and very quickly below 30 K down to 24 K (see Fig. 2). The fast increase of λ_{ZF} below 30 K is ascribed to the build of correlated fluctuations between the Ce magnetic moments in the paramagnetic phase. To check this, we have performed a field scan at 25 K in an applied longitudinal field (LF), parallel to the initial muon spin polarization. By doing this, at sufficiently large fields the contribution of the correlated fluctuations is suppressed, as well as that of the static nuclear-magnetic moments, and the μSR spectra is purely exponential. As one can see from the inset of Fig. 2, λ_{LF} is practically field independent for applied magnetic fields larger than 200 Oe. Moreover, $\lambda_{LF-1\,kOe}$ at 25 K (the down triangle in Fig. 2) equals λ_{ZF} (100 K), suggesting that the fluctuation rate is unchanged from 100 K down to at least 25 K.

Two well-defined oscillating signals (see the difference between the μSR spectra recorded at 22 and 23 K in Fig. 3) can be fitted for the μSR spectra recorded below 22 K, corresponding to muons stopped in two magnetically inequivalent crystallographic sites of ferromagnetic $CeIn_2$ binary alloy. The temperature dependence of these two frequencies, ν_1 and ν_2, corresponding to two oscillating signals below 22 K, is plotted in Fig. 4. For temperatures very close and above $T_C = 22$ K, for instance, 22.3 K, we already have a fast depolarized signal (that we had at 23 K) and some structure at the beginning of the spectra (quite similar to that which we had at 22 K), and the frequencies drop abruptly at the transition. From this, it is evident that the order parameter (magnetization) vanishes abruptly above 22 K, confirming that the ferromagnetic transition is of first order. The frequencies are proportional to the local field ($2\pi\nu = \gamma_\mu H_{\text{loc}}$) at the muon site, therefore the related values of H_{loc} change from

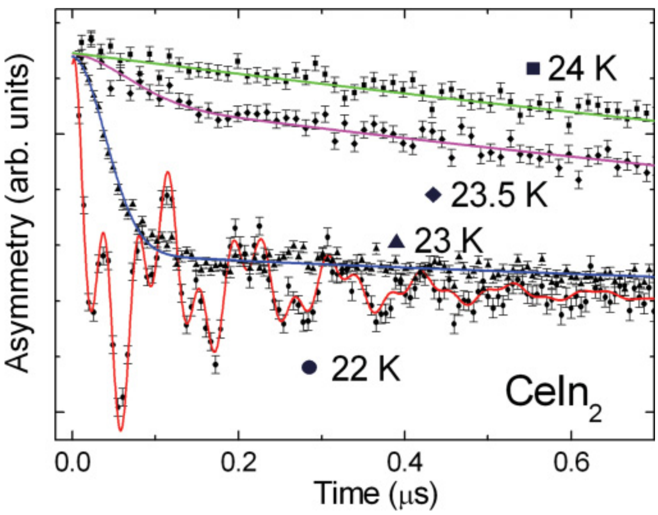

FIG. 3. (Color online) μSR spectra recorded for $CeIn_2$ at selected temperatures close to $T_C = 22$ K.

1.9 kOe at 22 K to 3.1 kOe at 10 K for ν_1, and from 0.7 to 1.08 kOe for ν_2, respectively. These values of the internal field H_{loc}, especially those related to ν_1, are relatively large when compared with those reported for other Ce, Yb, and U compounds,[11] and even larger than those of MnSi,[13] also with a first-order ferromagnetic transition. This may indicate a high degree of localization of $4f$ moments in $CeIn_2$, although H_{loc} does not only depend on the ordered moment, but also on the particularities of the crystallographic structure.

On the other hand, the μSR spectra recorded at temperatures between 22 and 24 K (see Fig. 3) show a fast depolarized signal at low times, indicating a large magnetic field distribution of static local fields at the muon site in $CeIn_2$ where even the first wiggle is heavily damped, resulting in no oscillation in the μSR spectra. This feature may be ascribed to incommensurate magnetic structures, some kind of disordered magnetism, or short-range order, where a monotonic relaxation of the muon spin polarization rather than a spin precession at defined frequencies is often observed.[14–16] Thus, from the μSR measurements, one of the possibilities we have is the existence of an additional magnetic phase, with incommensurate characteristics, above the first-order ferromagnetic transition ($T_C = 22$ K) in the $CeIn_2$ alloy. The presence of this phase could be somewhat hidden by the contribution of the ferromagnetic transition, as it was not clearly observed in previously reported specific-heat and dc(ac)-magnetic susceptibility data.[9] This result from μSR spectroscopy may be a relevant step toward understanding the first-order nature of ferromagnetism of the binary $CeIn_2$ alloy, since the crossover from paramagnetism to first-order ferromagnetism through an intermediate magnetic phase has been observed and very well documented in other Ce Alloys.[17,18] In order to support this result, a study of the evolution of the magnetic phases in the $La_{1-x}Ce_xIn_2$ series of alloys for high Ce concentrations ($0.9 \leqslant x \leqslant 1.0$), very close to that of the $CeIn_2$ alloy, is proposed in the next section. Similarly to that observed in other Yb and U systems,[10,19] the La substitution can promote a reduction in the ordering temperature, thereby allowing us to follow both the behavior of T_C and the ordering temperature of this eventual additional phase, which henceforth will be identified as T^*.

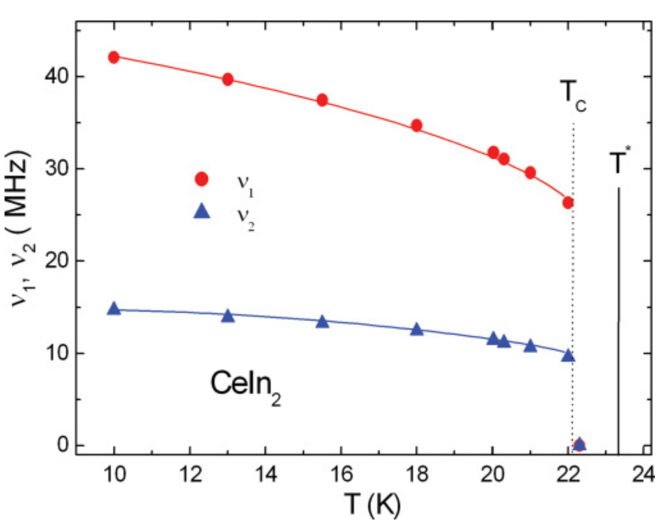

FIG. 4. (Color online) Temperature dependence of the zero field μ^+ frequencies measured in $CeIn_2$ below T_C. The lines are a guide to the eye. The frequencies abruptly vanish above 22 K, which is the sign of the first-order transition. T^* indicates the temperature at which the additional magnetic contribution appears.

B. Magnetic and thermodynamic properties of the $La_{1-x}Ce_xIn_2$ ($0.9 \leqslant x \leqslant 1.0$) series of alloys

We have started our study of the chemical diluted series with the $La_{0.05}Ce_{0.95}In_2$ alloy, with a Ce concentration very close to that of the pure $CeIn_2$ one. In Fig. 5, the temperature dependence of the in-phase component of the ac-magnetic susceptibility is depicted. The ferromagnetic transition shifts to a lower temperature than that of the parent alloy,[9] down to 20.8 K, and an additional faint anomaly is revealed at approximately $T^* = 22.4$ K. Also, the curves of *ac*-magnetic

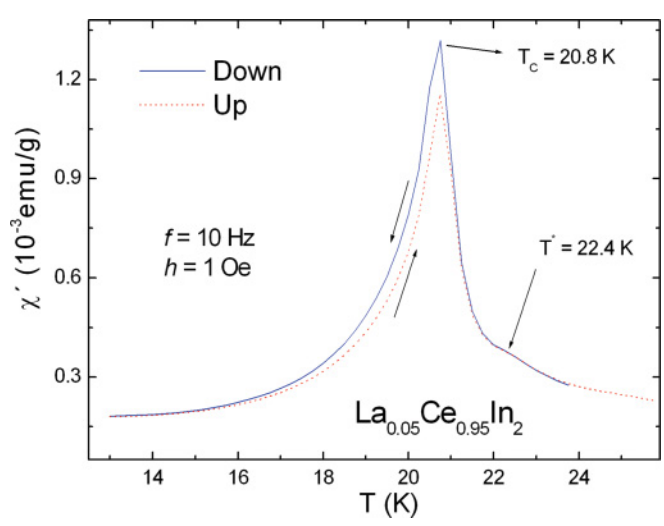

FIG. 5. (Color online) In-phase component of the AC susceptibility measured on warming and cooling in $La_{0.05}Ce_{0.95}In_2$ at a frequency of 10 Hz. It shows the thermal hysteresis characteristic of first-order transitions. The peak associated with the ferromagnetic (FM) transition shifts down to 20.8 K, in relation to the pure binary $CeIn_2$ alloy. An additional contribution around $T^* = 22.4$ K is revealed.

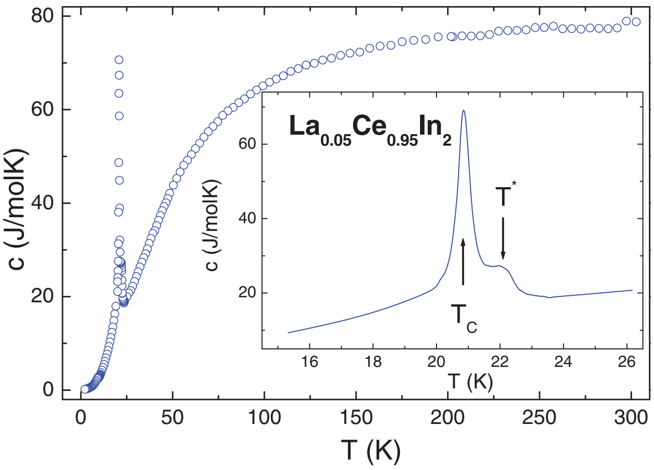

FIG. 6. (Color online) Temperature dependence of the specific heat of the $La_{0.05}Ce_{0.95}In_2$ alloy. The inset details the temperature range around the magnetic transition. Two transitions appear at T_C and T^*.

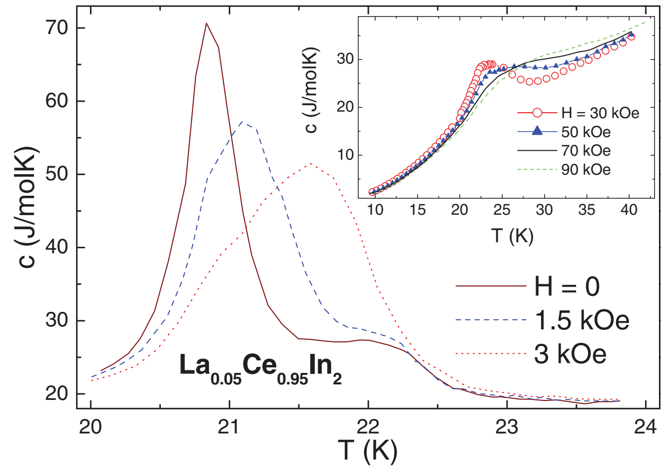

FIG. 7. (Color online) Specific heat in $La_{0.05}Ce_{0.95}In_2$ around the magnetic transitions as a function of the magnetic field. The contributions from the two transitions merge into a single one at 3 kOe. The inset shows the behavior for magnetic fields ranging from 30 kOe to 90 kOe.

susceptibility upon cooling and warming show a thermal hysteresis, similar to that observed in the $CeIn_2$ alloy, indicating the presence of metastable states, and therefore, that the ferromagnetic transition retains its first-order character despite the chemical substitution effects. These results are congruent with those obtained from the measurement of the specific heat on the same sample (see Fig. 6). A peak, sharper than that expected for a second-order transition, is observed at T_C. Furthermore, the specific-heat jump at the transition is of the order of 50 J/mol K. Although this value is smaller than the one found on the parent $CeIn_2$ compound, it is still much larger than the one expected for a second-order transition, 12.5 J/mol K. At this point, it is important to note that, as the specific-heat measurements have been carried out at a Quantum Design PPMS using the thermal relaxation method, the magnitude of the peak at the transition has certainly been underestimated. It has been shown that the default software used by this type of equipment does not allow us to accurately determine the real value of the specific heat at a first-order transition,[20] due to the fact that the relaxation technique measures over a finite temperature range, which, in our case, has been restricted to its minimum value when measuring around the transition. Thus, the magnitude of the specific-heat jump at the transition will be even higher, reinforcing the conclusion that the ferromagnetic transitions in both $CeIn_2$ (Ref. 9) and the La-diluted sample have a first-order character. When looking at the behavior around the transition in the $La_{0.05}Ce_{0.95}In_2$ alloy in greater detail (see the inset of Fig. 6), one notices an additional well-defined anomaly at temperatures slightly larger than T_C. This temperature matches the one already detected by the ac susceptibility, which was labeled as T^*. The shape of this new peak and the value of its specific-heat jump (~7 J/mol K) indicate the second-order character of this transition.

The field dependence of the specific heat around the magnetic transitions in the $La_{0.05}Ce_{0.95}In_2$ alloy is presented in Fig. 7. Increasing the magnetic field up to 1.5 kOe, the ferromagnetic transition shifts to higher temperatures, as expected, whereas this does not affect the position of the second anomaly at T^*. This indicates that the peaks do indeed correspond to different magnetic phases. The ferromagnetic transition is associated with a commensurate magnetic structure,[9] and the other one at T^* could be related to an incommensurate antiferromagnetic structure as suggested by the μSR spectroscopy results in the $CeIn_2$ alloy, and in a similar way as that which occurs in other Ce Alloys.[17,18] For an applied magnetic field of 3 kOe, both anomalies merge together into one single peak and, for higher magnetic fields, this peak broadens and shifts to higher temperatures, according to what is expected from a ferromagnetic behavior.[9]

The effect of the chemical substitution of Ce by La on the magnetic transition temperatures, T_C and T^*, is corroborated by the ac-magnetic susceptibility results on the $La_{0.1}Ce_{0.9}In_2$ alloy (Fig. 8). The ferromagnetic transition keeps on moving to

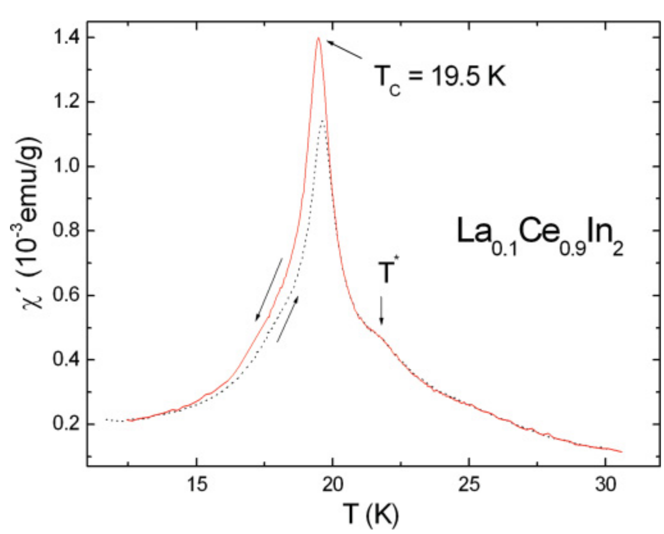

FIG. 8. (Color online) In-phase component of the ac susceptibility measured on warming and cooling in $La_{0.1}Ce_{0.9}In_2$ at a frequency of 10 Hz. It shows the thermal hysteresis characteristic of first-order transitions. An additional contribution at approximately 21 K is also observed.

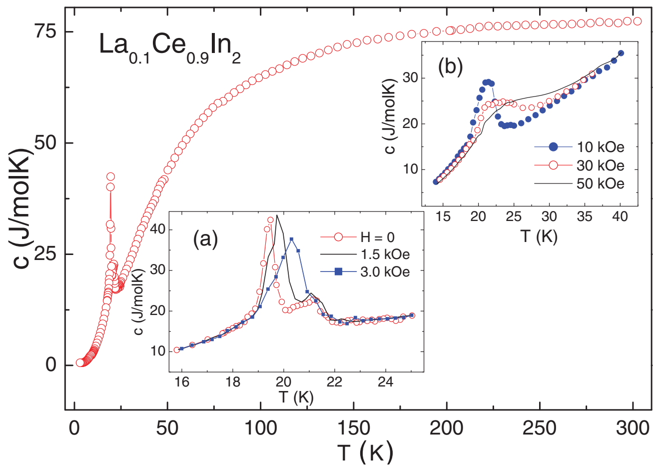

FIG. 9. (Color online) Temperature dependence of the specific heat of the $La_{0.1}Ce_{0.9}In_2$ alloy. The insets detail the field dependence of the magnetic contribution in different magnetic field ranges.

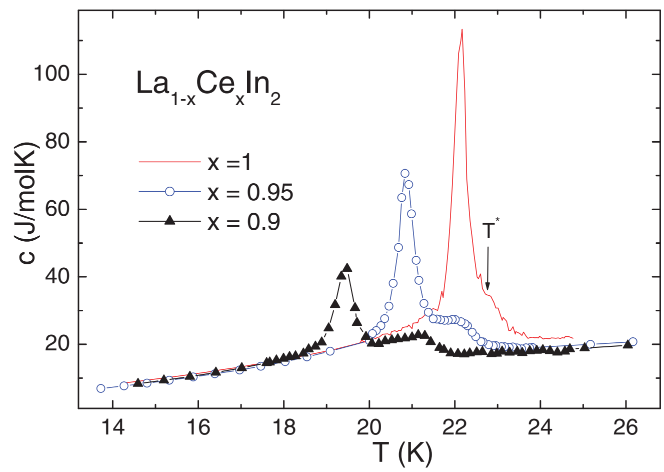

FIG. 10. (Color online) Temperature dependence of the specific heat for the $La_{1-x}Ce_xIn_2$ series of alloys with $x = 0.9$, $x = 0.95$, and $x = 1.0$. With the chemical substitution of Ce by La, the ferromagnetic transition shifts to lower temperatures, and an additional anomaly is revealed. This last anomaly (T^*) is conspicuously masked by the short-range correlations established above T_C in the $CeIn_2$ alloy ($x = 1.0$).

lower temperatures, in this case down to 19.5 K, and T^* moves down to 21 K. Measurements upon warming and cooling also show thermal hysteresis, indicating first-order features of the ferromagnetic transition. The ferromagnetic behavior at low temperatures in this alloy has been checked by measuring the hysteresis loop at 2.5 K (not shown). The remanence value ($M_r = 0.73\ \mu_B$/Ce) is similar to that reported in the $CeIn_2$ alloy, whereas the coercivity value (3.6 kOe) is larger, which may reflect a higher magnetocrystalline anisotropy and/or the influence of disorder effects, creating more pinning sites within the ferromagnetic domains. The specific-heat measurements performed on the $La_{0.1}Ce_{0.9}In_2$ alloy are presented in Fig. 9. Although there is a clear reduction of the specific-heat jump at T_C, not fully justified by the smaller Ce content, the height of this jump (~30 J/mol K), which can be larger considering the necessary correction in the measurements as commented above, is still enough to certify the first-order character of the transition. Inset (a) of Fig. 9 shows that the transition at T^* is still present at temperatures larger than T_C and that the La dilution moves both transitions to lower temperatures, in agreement with the ac-susceptibility results. This inset also shows that the increase of the applied magnetic field shifts T_C to higher temperatures until it merges with T^*. Once both anomalies have joined [see inset (b) of Fig. 9], the further increase of the magnetic field shows a typical ferromagnetic behavior.

IV. DISCUSSION

As mentioned above, the μSR spectra recorded between 22 and 24 K (see Fig. 3) show a fast depolarized signal at low times in $CeIn_2$, with a broad distribution of static local fields at the muon site, characteristic of incommensurate magnetic structures.[11,14,16] In fact, the chemical substitution of Ce by La reveals an additional contribution at T^*, above T_C, in the ac-magnetic susceptibility and specific heat for the samples with a Ce concentration of $x = 0.9$ and $x = 0.95$. Furthermore, these results indicate that the magnetic transition taking place at T^* is closer to T_C in $CeIn_2$ than in the La-diluted alloys with $x = 0.9$ and $x = 0.95$, and thus it might be masked by the short-range magnetic correlations established above T_C, as can be concluded from Fig. 10.

Useful information concerning the magnetic transitions can also be extracted from the behavior of the magnetic entropy (S_{mag}). S_{mag} can be calculated from the expression $S_{mag} = \int (c_{mag}/T)dT$, where c_{mag} is the magnetic contribution to the specific heat, obtained with the same procedure as the one used on the $CeIn_2$ alloy.[9] The two magnetic transitions at T_C and T^* in the $La_{0.1}Ce_{0.9}In_2$ alloy can be clearly identified from the temperature dependence of S_{mag} in Fig. 11. In the case of the $CeIn_2$ alloy, both transitions are not distinguished because the corresponding anomalies are merged together.[9] In addition, as already reported,[9] a steep decrease at T_C is

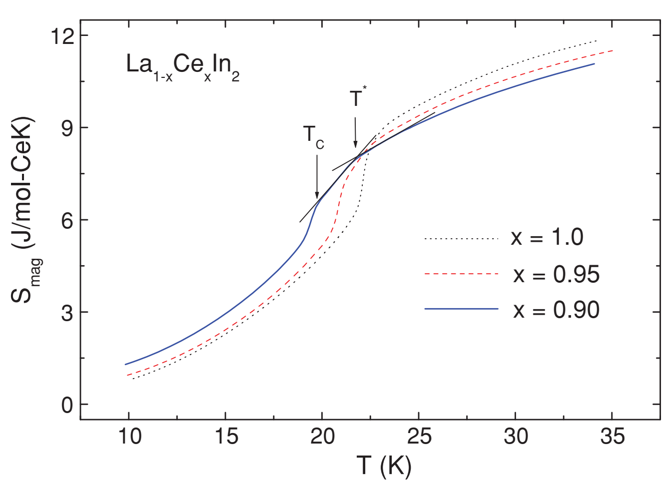

FIG. 11. (Color online) Temperature dependence of the magnetic entropy (S_{mag}) in the $La_{1-x}Ce_xIn_2$ series of alloys with x ranging from 0.9 to 1.0. The steep decrease at the magnetic transition softens with the chemical substitution of Ce by La. Two distinct transitions can be identified for $x = 0.9$.

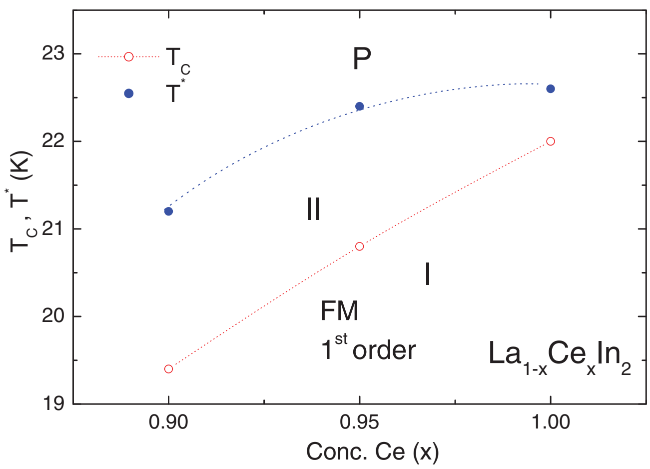

FIG. 12. (Color online) Phase diagram of the $La_{1-x}Ce_xIn_2$ series of alloys as a function of the Ce concentration (x). An additional magnetic phase appears in region II, between the paramagnetic (P) and ferromagnetic (FM) phases. The lines are just a guide for the eye.

observed in the $CeIn_2$ alloy ($x = 1$), although it is somehow smoothed with respect to the ideal system, as expected from the polycrystalline nature of the sample.[21] However, it is clearly much steeper than the one found in the alloys with $x = 0.9$ and $x = 0.95$, showing a partial rounding. This result can be easily understood, as it is well known that thermally driven first-order transitions may be rounded by quenched disorder, which means that a first-order transition in a pure system can gradually become continuous upon doping.[21] Despite these disorder effects on the first-order transition due to the chemical substitution process, the study of the Ce substitution by La in the $CeIn_2$ alloy has achieved the goal of splitting the two existing transitions on this alloy, and thus enlightening the mechanism that rules such a peculiar first-order transition.

The phase diagram of Fig. 12 shows the existence of a region (labeled as II), associated with an intermediate magnetic phase between the paramagnetic and first-order ferromagnetic ones. This kind of intermediate state has been postulated, for instance, in the study of the so-called Lifshitz point,[22] and/or in the anisotropic next-nearest-neighbor Ising (ANNNI) model.[23] The presence of this additional magnetic precursor has also been observed in the Ce_3Bi_4 intermetallic, with a first-order transition from an incommensurate magnetic phase to a ferromagnetic phase at 3.3 K ($\Delta c_{mag} \approx 18$ J/mol CeK),[17] and in the $CeRu_2Ge_2$ alloy, with a double transition, from a paramagnetic state into an antiferromagnetic one ($T_N \approx 8.3$ K) and then into a ferromagnetic one ($T_C \approx 7.5$ K, $\Delta c_{mag} \approx 45$ J/mol K).[18] Although both the values of $T_C = 22$ K and $\Delta c_{mag} = 103$ J/mol K (can be larger), obtained at the transition in $CeIn_2$, are enhanced with respect to those other Ce alloys, the mechanism allowing the existence of a first-order ferromagnetic transition is similar, involving the crossover from paramagnetism to ferromagnetism through an intermediate magnetic phase. This intermediate magnetic phase in $CeIn_2$ has been unveiled from the chemical substitution study and supported by the μSR experiments, in good agreement with the above reported models.[22,23] Concerning the nature of the magnetic transition at T^*, the experimental evidences in the other (cited above) Ce systems point toward a transition into an incommensurate modulated antiferromagnetic structure.[17,18]

V. CONCLUSIONS

The μSR measurements in $CeIn_2$ have indicated the existence of an additional magnetic contribution above T_C, between 22 and 24 K. This is supported by the study of the magnetic and thermal properties of the $La_{1-x}Ce_xIn_2$ series of alloys ($0.9 \leqslant x \leqslant 1.0$), revealing that this contribution is related with a long-range ordering rather than a short-range one. So far, these results show the intrinsic nature of the first-order ferromagnetism in the binary $CeIn_2$ alloy, as a crossover from the paramagnetic state into an intermediate magnetic phase, through a second-order phase transition, and finally, to a ferromagnetic state, through a first-order phase transition, in a similar way as observed in other Ce systems.[17,18] Finally, it would be essential to carry out pressure experiments in both the $CeIn_2$ sample and the La-substituted series of alloys, in order to determine the evolution of the different magnetic phases.

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