# Development of the heavy-fermion state in $Ce_2IrIn_8$ and the effects of Ce dilution in $(Ce_{1-x}La_x)_2IrIn_8$

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We report a study of muon Knight shifts to investigate the formation of the heavy-fermion state in single crystals of $(Ce_{1-x}La_x)_2IrIn_8$. Two different kinds of Knight-shift anomalies (deviations from a linear relation between the Knight shift and the susceptibility) are found: (1) a high-temperature effect arising from depopulation of crystalline electric field levels with temperature, and (2) a lower-temperature anomaly arising from the onset of the heavy-fermion state below a characteristic temperature $T^*$, in agreement with the "two-fluid" model of heavy-fermion formation. In $Ce_2IrIn_8$, we find $T_c^*=20.0(6)$ K and $T_a^*=15.2(1.2)$ K for applied field $H \parallel c$ axis and $H \parallel a$ axis, respectively. For the Ce diluted systems $(Ce_{1-x}La_x)_2IrIn_8$, $x=0.1$, 0.25, 0.50, 0.70, and 0.90, $T^*$ decreases linearly for $x \leq 0.5$, reaching zero near $x=0.7$, indicating the reduction in intersite $f$-spin correlations with Ce dilution. A comparison with nuclear magnetic-resonance measurements of Knight-shift anomalies in several other heavy-fermion compounds suggests that the observed small anisotropy in $T^*$ may be induced by the applied field, and that $T^*$ may be inherently isotropic, even in highly anisotropic materials.

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## I. INTRODUCTION

A key characteristic of $f$-electron heavy-fermion compounds is the observation of a crossover from localized $f$-electron behavior at high temperatures, where the $f$ electrons scatter the conduction electrons independently as local impurities, to the formation of a Kondo liquid of interacting $f$ electrons at low temperatures, where coherent conduction-electron scattering occurs. Typically, this behavior has been observed as a broad maximum in the resistivity. Although the single-site impurity problem, i.e., the Kondo effect, has been solved exactly,[1] a general solution for the Kondo lattice has not been found because of complex intersite coupling effects. Recently, however, a semiquantitative model has been proposed which significantly improves our understanding of the relation between the single-site Kondo effect and the formation of a Kondo liquid in the presence of intersite coupling.[2] In this light it is important to provide new data in a variety of heavy-fermion materials to test these ideas.

Knight-shift anomalies, which are deviations from a linear relation between the Knight-shift $K$ and the bulk susceptibility $\chi$, are often observed at low temperatures in heavy-fermion materials. Generally, $K$ is expressed as $K = A\chi/(N_A\mu_B)$, where $A$ is the hyperfine coupling constant, $N_A$ is Avogadro's number and $\mu_B$ is the Bohr magneton. Possible reasons for a nonlinear $K$-$\chi$ relation can involve a change in the hyperfine coupling-constant $A$ with temperature or the onset of an additional component of $\chi$, or both concurrently. An example of the former involves the depopulation of low-lying $f$-electron crystalline-electric-field (CEF) levels,[3,4] which can change $A$. A recently-introduced example of the latter is the so-called two-fluid model of heavy-fermion formation.[2,5–8] Here, in addition to the high-temperature $\chi$ representing the local $f$ moments, a low-temperature susceptibility component $\chi_{KL}$, characteristic of the coherent Kondo-liquid (KL) state below temperature $T^*$, is introduced. This phenomenological model was first discussed by Nakatsuji *et al.*,[5,6] who found scaling behavior through a careful analysis of specific heat and magnetization data in $Ce_{1-x}La_xCoIn_5$ and $CeIrIn_5$. Curro *et al.*[7] showed that this two-fluid model seems to provide the explanation for many nuclear magnetic-resonance (NMR) Knight-shift anomalies observed in a variety of Ce-, Yb- and U-based heavy-fermion compounds. Yang and Pines[8] derived a new scaling relation for the temperature dependence of $\chi_{KL}$, which deviates somewhat from the original expression given in Nakatsuji *et al.*[5,6] Yang and Pines showed that the low-temperature KL possesses a non-Landau density of states at the Fermi surface below $T^*$ varying as $(1-T/T^*)^{3/2}[1+\ln(T^*/T)]$. Moreover, this KL density of states was found to be in excellent agreement with microscopic calculations of the density of states in $CeIrIn_5$ using dynamic mean-field theory.[9]

$Ce_2MIn_8$ is a bilayer variant of $CeMIn_5$ ($M$=Co, Rh, Ir) with a high Sommerfeld constant of $\gamma \sim 700$ mJ/mol-Ce $K^2$.[10] Resistivity measurements show a broad maximum at around 50 K, suggesting the onset of coherent state at lower temperatures.[11] The magnetic, zero-field specific-heat $C/T$ of $Ce_2IrIn_8$, measured in the temperature range $\cong 0.4-8$ K, is approximately constant between 5 and 8 K, rising dramatically for $T < 5$ K.[12] This material was previously investigated by muon spin relaxation ($\mu$SR) and found to undergo small-moment disordered spin freezing below $T_g=0.6$ K.[13]

In this paper we present muon Knight-shift measurements in $(Ce_{1-x}La_x)_2IrIn_8$ undertaken to investigate the general applicability of the two-fluid model in highly anisotropic heavy-fermion compounds, and to study the effects of Ce dilution on the local magnetic susceptibility measured at several interstitial muon sites in the lattice. A key question concerns the degree to which $T^*$ varies with the direction of the applied magnetic field, given the anisotropy of the bulk susceptibility. A preliminary report of muon Knight-shift results in $Ce_2IrIn_8$ was presented earlier,[14] but only for experiments in applied fields of 1 T. The detailed study presented here extends experiments to La-doped samples, to applied fields of up to 3 T, and determines the full angular dependence of the Knight shifts, allowing assignment of the muon sites and correcting errors in the preliminary report.[14]

We emphasize that one must carefully distinguish between Knight-shift anomalies originating from CEF and heavy-fermion effects in order to properly interpret any underlying two-fluid heavy-fermion physics. We find that the temperature dependence of the two-fluid anomalies in $(Ce_{1-x}La_x)_2IrIn_8$ scales with $T^*/T$ in accordance with the scaling relation of Yang and Pines noted above. Furthermore, the characteristic temperature $T^*$ for the intersite interactions in $Ce_2IrIn_8$ is nearly independent of the direction of the applied magnetic field in these tetragonal materials. In $(Ce_{1-x}La_x)_2IrIn_8$, $T^*$ decreases approximately linearly with $x$ for $x \leq 0.5$, extrapolating to zero near $x=0.7$, indicating the limit to which the $f$-electrons can be diluted and still enter a coherent Kondo liquid state.

## II. EXPERIMENTAL DETAILS

Single-crystal samples were grown by the In-flux method at Los Alamos National Laboratory.[15] Two sets of $Ce_2IrIn_8$ single crystals were prepared, one for $H \parallel c$ and the other for $H \parallel a$. Five different single crystals of $(Ce_{1-x}La_x)_2IrIn_8$ for $H \parallel c$ $(x=0.1, 0.25, 0.5, 0.7, 0.9)$ were also grown. Transverse-field (TF) $\mu$SR experiments with the applied magnetic field perpendicular to the muon spin were performed at the M20 (Helios spectrometer) and M15 (HiTime spectrometer) beamlines at TRIUMF in Vancouver, Canada. The samples were mounted on silver plates, which served as a reference material to calibrate the applied field. In the HiTime spectrometer a muon veto-counter-system was used to separately collect positron events from muons which either stopped in the sample or missed it. Knight shifts were measured with $H=1-3$ T, and susceptibility measurements were performed in the corresponding fields on the same samples as used for the $\mu$SR measurements.

The TF-$\mu$SR spectra were fit to a sum of exponentially damped cosine functions corresponding to muons stopping at different crystalline lattice sites and a background silver signal. The observed muon Knight shift is defined as $K_{\mathrm{exp},i} = (\omega_i - \omega)/\omega = K_{\mu,i} + K_{\mathrm{dem}}$, where $\omega_i/2\pi$ is the measured precession frequency of the $i$-th signal component, $\omega = \gamma_\mu H$ is the muon reference frequency in the external field $H$, $\gamma_\mu$ is the muon gyromagnetic ratio $(=2\pi \times 135.54$ MHz/T) and $K_{\mathrm{dem}} = 4\pi(1/3 - N)\rho_{\mathrm{mol}}\chi$ is the contribution from the Lorentz and demagnetization terms. Here $N$ is the demagnetization factor for a particular sample-field orientation and $\rho_{\mathrm{mol}}$ is the molar density.

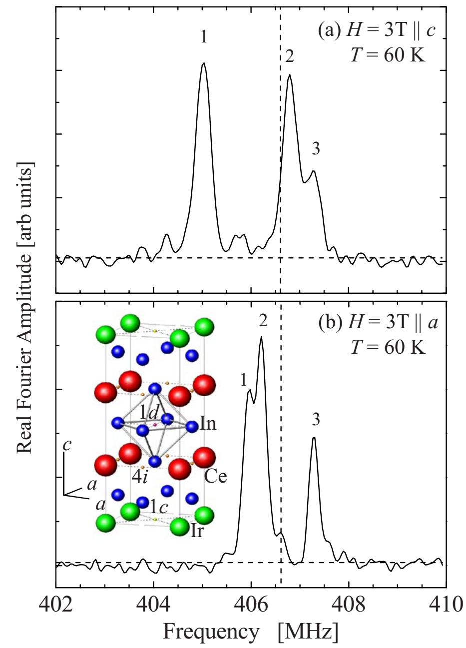


FIG. 1. (Color online) The fast Fourier transform spectra of $\mu$SR signals under $H=3$ T at 60 K with (a) $H \parallel c$ and (b) $H \parallel a$ in $Ce_2IrIn_8$. The dashed line shows the $K=0$ frequency. The inset shows the crystal structure of $Ce_2IrIn_8$. Wyckoff positions for the $1c$, $1d$, and $4i$ sites are noted.

Following the convention of Yang and Pines,[8] the uniform susceptibility can be written as $\chi(T) = f(T)\chi_{KL} + [1 - f(T)]\chi_{SL}$, where $\chi_{KL}$ and $\chi_{SL}$ are the susceptibilities of the Kondo Fermi liquid, which sets in below $T^*$, and the high-temperature spin liquid, respectively. The relative fraction $f(T)$ lies between 0 and 1. The muon Knight shift from the $i$-th frequency component, including the contribution from $\chi_{KL}$, is summarized as follows:[7]

$$K_{\mu,i} = K_{\mathrm{exp},i} - K_{\mathrm{dem}} \tag{1}$$

$$= K_0 + [A_i f(T)\chi_{KL} + B_i[1 - f(T)]\chi_{SL}]/(N_A\mu_B), \tag{2}$$

where $K_0$ is a temperature-independent shift. The spin-liquid component is coupled to the muon by hyperfine coupling-constant $B_i$, while the itinerant KL component is coupled by hyperfine coupling $A_i$. For muons these hyperfine couplings are typically the sum of a contact term from the conduction electrons and a dipolar term from the $f$ electrons. The Knight-shift anomaly from the KL can be written as[7,8]

$$K_{\mathrm{anom}} = K_\mu - K_0 - B\chi/(N_A\mu_B) = (A - B)f(T)\chi_{KL}/(N_A\mu_B) \tag{3}$$

$$= K^0_{\mathrm{anom}}(1 - T/T^*)^{3/2}[1 + \ln(T^*/T)]. \tag{4}$$

We note that $K_{\mathrm{anom}} = K^0_{\mathrm{anom}}$ for $T/T^* \simeq 0.369$.

## III. DATA ANALYSIS AND RESULTS

### A. $Ce_2IrIn_8$

Figure 1 shows the fast Fourier transform (FFT) spectra from the HiTime spectrometer at 60 K with $H=3$ T parallel

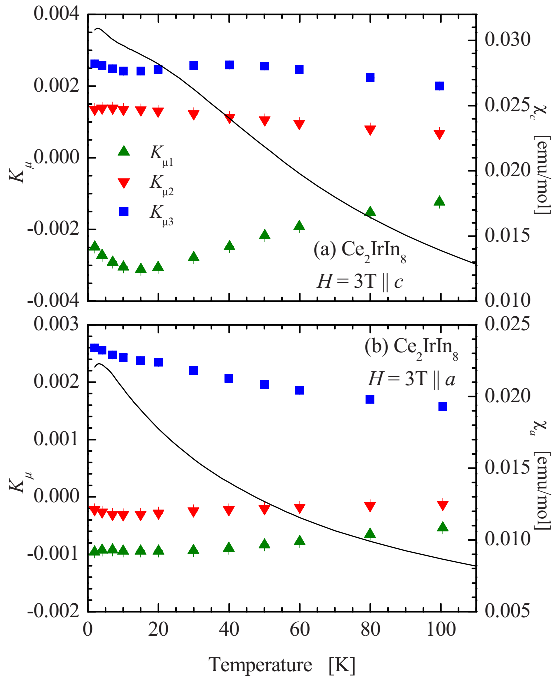


FIG. 2. (Color online) Temperature dependence of muon Knight-shift $K_\mu$ and bulk susceptibility $\chi$ in Ce$_2$IrIn$_8$ for $H=3$ T: (a) $H\|c$ and (b) $H\|a$.

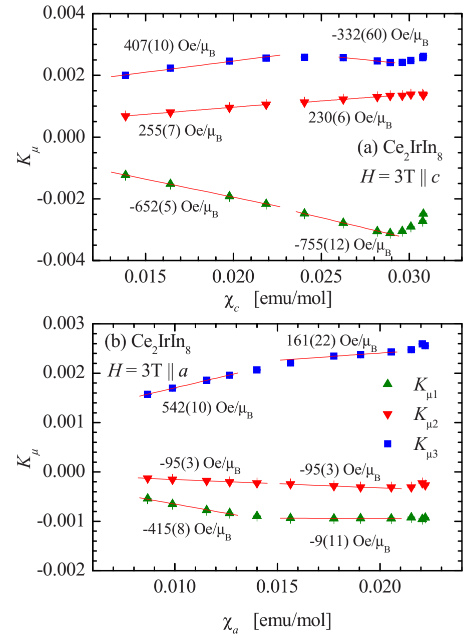


FIG. 3. (Color online) $K_\mu$ versus $\chi$ with (a) $H\|c$ and (b) $H\|a$ in Ce$_2$IrIn$_8$.

to the $c$ and $a$ axes in Ce$_2$IrIn$_8$. The dashed line shows the $K=0$ frequency taken in a separate histogram. It is apparent that three shifts (labeled 1, 2, and 3 from the lowest frequency) were observed for both field directions. Figure 2 shows the temperature dependence of $K_\mu$ and the susceptibility in Ce$_2$IrIn$_8$ for $H\|c$ and $H\|a$ with $H=3$ T. The $K_\mu$ versus $\chi$ plots are shown in Fig. 3. Note the behavior of $K_{\mu,3c}$: there are two breaks from linearity around 0.024 and 0.029 emu/mol in Fig. 3, corresponding to temperatures ~50 and ~20 K as shown in Fig. 2, respectively. A change in linear slope around 0.024 emu/mol is also observed in $K_{\mu,1c}$ and $K_{\mu,2c}$ for $H\|c$. Similar behavior is seen in the $H\|a$ data around 0.014 emu/mol (~50 K) in $K_{\mu,1a}$ and $K_{\mu,3a}$; this tendency is not as clear for $K_{\mu,2a}$ probably because of the small value of the shift. As discussed below, we assert that the higher-temperature Knight-shift anomalies near $T=50$ K are due to CEF effects, and the lower temperature anomalies to heavy-fermion physics. Quantitative estimates of the $B_i$ values corresponding to linear $K$-$\chi$ relations above and below $T=50$ K were obtained from Eq. (2) by setting $f(T)\equiv 0$. These values are shown next to the curves in Fig. 3. The $K_0$ values are shown in Table I. Before discussing the Knight-shift anomalies in more detail, we first turn to a determination of the probable muon lattice sites.

The relative asymmetries for the three signals were determined from fits to the $\mu$SR precession signals corresponding to the shifts in Fig. 3: Asy$_{1c}$:Asy$_{2c}$:Asy$_{3c}$=48%:37%:15% and Asy$_{1a}$:Asy$_{2a}$:Asy$_{3a}$=25%:52%:23% for $H\|c$ and $H\|a$, respectively. Furthermore, data were taken in the Helios and HiTime spectrometers at $H=2$ T as a function of the direction of the applied magnetic field. Figure 4 shows the angular dependence of the Larmor precession frequencies when $H$ is turned in the $(a,c)$ plane. Here $\theta=0°$ and 90° correspond to $H\|a$ and $H\|c$, respectively. Note that the asymmetries are independent of $\theta$.

There are three likely candidates for the muon sites, namely, the 1$c$, 1$d$, and 4$i$ Wyckoff positions, as shown in Fig. 1(b). A fourth Wyckoff site, the 1$b$ site, which sits between two Ce atoms [at (0,0,0.5)], is ruled out because the calculated dipole field at this position (2.46 kOe/$\mu_B$) is much higher than any of the measured $B_i$ values. The 1$c$ and 1$d$ sites are axially symmetric, which means that $B_i=B_{c,i}+B^{zz}_{\mathrm{dip},i}$ for $H\|c$ and $B_i=B_{c,i}-\frac{1}{2}B^{zz}_{\mathrm{dip},i}$ for $H\|a$. Here $B^{zz}_{\mathrm{dip},i}$ is the $z$ component of the dipole coupling tensor. The contact field $B_{c,i}$ is assumed to be isotropic. The 4$i$ sites comprise two magnetically inequivalent, equally occupied, subsites yielding two frequencies when $H\|a$. These subsites become equivalent when $H\|c$, producing a single frequency with twice the asymmetry.

The site assignments are shown in Fig. 4. This determination is confirmed both from the relative asymmetries and from a comparison between the extracted values of the dipolar fields $B^{zz}_{\mathrm{dip},i}$ at the axially symmetric 1$c$ and 1$d$ sites and their calculated values. We use the $K_{\mu,2c}$, $K_{\mu,3c}$, and $K_{\mu,2a}$ shifts, the latter comprising the sum of $B_i=B_{c,i}-\frac{1}{2}B^{zz}_{\mathrm{dip},i}$, $i=1c$, and 1$d$, weighted by their respective asymmetries. Thus, $K_{\mu,3c}=B_{c,1d}+B^{zz}_{\mathrm{dip},1d}$, $K_{\mu,2a}=\alpha(B_{c,1c}-\frac{1}{2}B^{zz}_{\mathrm{dip},1c})+\beta(B_{c,1d}-\frac{1}{2}B^{zz}_{\mathrm{dip},1d})$, and $K_{\mu,2c}=B_{c,1c}+B^{zz}_{\mathrm{dip},1c}$, where $\alpha=37/52$ and $\beta=15/52$ are the relative asymmetries. One obtains $\alpha B^{zz}_{\mathrm{dip},1c}+\beta B^{zz}_{\mathrm{dip},1d}=264$ Oe/$\mu_B$, which compares well with the calculated dipole fields $B^{zz}_{\mathrm{dip},1c}=428$ Oe/$\mu_B$ and $B^{zz}_{\mathrm{dip},1d}=-151$ Oe/$\mu_B$, yielding a weighted sum of 261 Oe/$\mu_B$. Using the calculated individual dipole fields one can then obtain $B_{c,1c}=-173$ Oe/$\mu_B$ and $B_{c,1d}=558$ Oe/$\mu_B$. The 4$i$ site does not possess axial symmetry and, thus, the data do not allow an easy determination of the dipole and contact fields.

As shown in Fig. 3, a linear $K$-$\chi$ relation holds at high temperatures, and there is a break in this linear slope at

TABLE I. Parameters $K_0$ and $K^0_{anom}$ derived from comparing all muon Knight-shifts $K_\mu$ in $Ce_2IrIn_8$ to Eqs. (2) and (4).

| Parameter | $K_{\mu,1c}$ | $K_{\mu,2c}$ | $K_{\mu,3c}$ | $K_{\mu,1a}$ | $K_{\mu,2a}$ | $K_{\mu,3a}$ |
|---|---|---|---|---|---|---|
| $K_0$ | 0.00039(2) | 0.000053(6) | 0.0010(1) | 0.00009(5) | 0.000016(5) | 0.00073(2) |
| $K^0_{anom}$ | 0.00034(9) | | 0.00012(1) | | 0.00005(4) | 0.00006(7) |

~50 K for all six shifts. This behavior—a change in the linear $K$-$\chi$ slope—is similar to that observed in $CeCu_2Si_2$ NMR measurements,[4] where it was argued that the change in slope occurs due to a depopulation of the CEF levels as the temperature is lowered, changing the effective hyperfine coupling. This is to be contrasted with the clear nonlinear behavior seen in the $K$-$\chi$ curve for $K_{1c}$ above $\chi$ =0.029 emu/mol (~20 K). A similar low-temperature break is seen clearly in the $K_{3c}$ curve, and in the $K_{3a}$ curve above $\chi$=0.021 emu/mol (~20 K). We, therefore, tentatively attribute the higher-temperature anomalies at $T_{CEF}$ ≅50 K in $Ce_2IrIn_8$ to CEF effects, and the lower-temperature anomalies to heavy-fermion effects described by Eqs. (3) and (4). This assignment is discussed further in Sec. IV below.

To extract values of $K_{anom}$ we use the values of $B_i$ obtained from linear fits to $K$ versus $\chi$ in the temperature region between about 25–50 K and employ Eq. (3). Figure 5(a) shows the temperature dependence of $K_{anom}$ for four of the shifts. No measurable anomaly could be determined for $K_{\mu,2c}$ and $K_{\mu,1a}$, indicating that the hyperfine coupling term $A-B$ in Eq. (3) is nearly zero. The data in Fig. 5(a) are well-described by Eq. (4) with $T_c^*$=20.0(6) K and $T_a^*$ =15.2(1.2) K for $H \parallel c$ and $H \parallel a$, respectively. The values of $K^0_{anom}$ are shown in Table I.

### B. $(Ce_{1-x}La_x)_2IrIn_8$

The effects of Ce dilution on the formation of heavy-electron state were investigated in La-doped samples for $H \parallel c$ in the temperature range between 65 and 2 K. (A preliminary result for $x$=0.1 and $H$=1 T was reported previously).[16] Figure 6 shows the FFT spectra in $(Ce_{1-x}La_x)_2IrIn_8$ at $T$ =60 K. One sees that La-doping induces a splitting of the lines seen in the undoped sample. This is most clearly exhibited by the lowest-frequency shift $K_{\mu,1c}$, which is cleanly separated from higher-frequency lines in the $x$=0 sample. $K_{\mu,1c}$ splits into two lines, the original line and a satellite line $K'_{\mu,1c}$ at somewhat higher frequency, which increases in intensity and shifts to higher frequency (smaller absolute shift) as $x$ increases. These two lowest-frequency lines from the 4$i$ site appear to represent the cleanest cases to study, in contrast to the higher-frequency 1$c$ and 1$d$ lines, where the shifts are smaller in magnitude and the satellite shifts likely overlap with the unsplit shifts. We, therefore, focus on the $K_{\mu,1c}$ and $K'_{\mu,1c}$ lines, noting that for $x$=0.75 and $x$=0.90 two separate lines cannot be distinguished.

The 4$i$ site possesses two nearest-neighbor Ce sites and four Ce sites in the next-nearest-neighbor positions. We assume that a muon site whose nearby atomic sites are depleted of Ce will exhibit a smaller absolute shift than if the atomic sites are Ce-rich. If the La goes into the lattice completely randomly (i.e., no chemical clustering), then the probability that the La occupies one or more of these sites should obey a binomial distribution. Experimentally, this probability is given by the relative asymmetries for the two lines. The measured relative percent asymmetries of $K_{\mu,1c}$:$K'_{\mu,1c}$ are as follows: 90:10 ($x$=0.1), 64:36 ($x$=0.25),

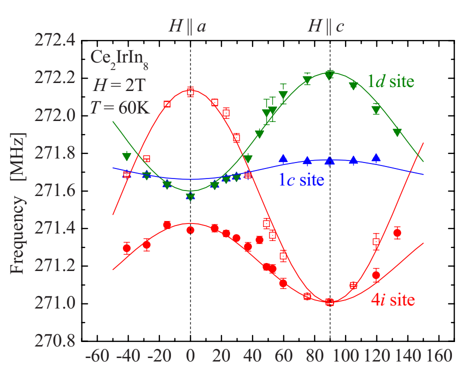


FIG. 4. (Color online) Angular dependence of the Larmor precession frequencies in $Ce_2IrIn_8$ with $H$=2 T. $H$ is turned in the $(a,c)$ plane.

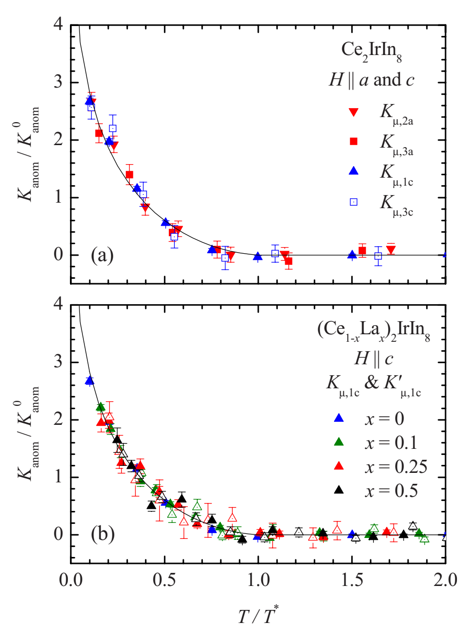


FIG. 5. (Color online) Temperature dependence of the normalized Knight-shift $K_{anom}/K^0_{anom}$ in (a) $Ce_2IrIn_8$ and (b) $(Ce_{1-x}La_x)_2IrIn_8$. The markers of solid and open triangles stand for $K_{\mu,1c}$ and $K'_{\mu,1c}$, respectively. Solid lines are fits to Eq. (4).

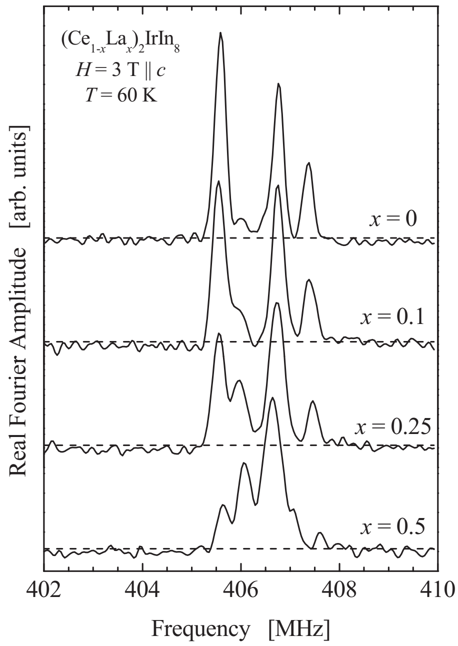


FIG. 6. The fast Fourier transform spectra of $\mu$SR signals under $H=3$ T at 60 K in the various La-doped samples.

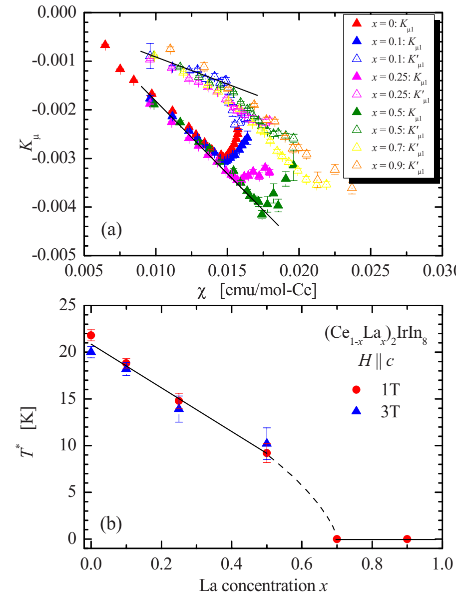


FIG. 7. (Color online) (a) $K_\mu$ versus $\chi$ plot in $(Ce_{1-x}La_x)_2IrIn_8$, where $\chi$ is normalized per Ce atom, and (b) La concentration dependence of $T^*$ in $(Ce_{1-x}La_x)_2IrIn_8$. The solid and dotted lines are guides for the eyes.

and 28:72 ($x=0.50$). (Here we have averaged the $H=1$ T and $H=3$ T data.) The binomial distribution for having either 0 or 1 La in any of four Ce sites, compared to having $\geq 2$ La in these sites, yields 95:5 ($x=0.1$), 74:26 ($x=0.25$), and 31:69 ($x=0.50$). Similarly, for two nearest-neighbor Ce atoms the binomial distribution predicts the ratios 81:19 ($x=0.1$), 56:44 ($x=0.25$), and 25:75 ($x=0.5$) for having 0 compared to $\geq 1$ La in the (nearest-neighbor) sites. Although the correlation between the data and these crude estimates is only semiquantitative (probably due to nonstatistical clustering), we conclude that the $K_{\mu,1c}$ line corresponds to a local Ce-rich region and the $K'_{\mu,1c}$ to a local Ce-depleted region.

A plot of $K$ versus $\chi$ for the La-doped materials shows similar behavior to the undoped system. Because data in $(Ce_{1-x}La_x)_2IrIn_8$ for $x>0$ were only accumulated up to $T=60$ K, the break in linearity near $T_{CEF}=50$ K found for $x=0$ is more difficult to discern, especially in the $x=0.1$ material. Nevertheless, a similar weak break is found near $T_{CEF}=40$ K for $x=0.25$ and near $T_{CEF}=25$ K for $x=0.50$ in both the $K_{\mu,1c}$ and $K'_{\mu,1c}$ shifts. We attribute these observations to a changes in the CEF splittings with overall La doping.

When $\chi$ is normalized per Ce atom, the high-temperature $K_{\mu,1c}$ shifts fall more or less onto a single $K$-$\chi$(emu/mol-Ce) line, revealing very clearly the low-temperature $T^*$ anomalies as deviations from this line for $x \leq 0.5$, as seen in Fig. 7(a). The $K'_{\mu,1c}$ shifts for $0.10 \leq x \leq 0.50$ show similar $T^*$ anomalies, as well as rough scaling with the Ce concentration for $T \gg T^*$, but with smaller negative high-temperature slopes than found for $K_{\mu,1c}$ [Fig. 7(a)], corresponding to smaller hyperfine fields.

The anomalous Knight-shifts $K_{anom,i}(i=1,1')$ for the $x \leq 0.50$ can be derived as before, and the $T^*$ values extracted as a function of La doping. This yields $T^*=18.2(2)$ K ($x=0.1$), 13.9(1.4) K ($x=0.25$) and 10.2(1.7) K ($x=0.5$), shown in Fig. 5(b), which shows the scaling behavior for $K_{anom,i}(i=1,1')$, $x \leq 0.50$, and in Fig. 7(b), which shows $T^*$ as a function of $x$. The values of $K_0$ and $K^0_{anom}$ in the La-doped system are summarized in Table II.

## IV. DISCUSSION AND CONCLUSIONS

The data presented here for $(Ce_{1-x}La_x)_2IrIn_8$ show that Knight-shift anomalies (deviations from a linear $K$ versus $\chi$ behavior) are a useful probe to study changes in the electronic correlations in heavy-fermion systems. This was noted previously by Curro *et al.*[7] In this paper we emphasize that to interpret these anomalies correctly, however, one must take full account of the several possible sources of nonlinear $K$-$\chi$

TABLE II. Parameters $K_0$ and $K^0_{anom}$ derived from comparing muon Knight-shifts $K_{\mu,1c}$ and $K'_{\mu,1c}$ in $(Ce_{1-x}La_x)_2IrIn_8$ ($0.1 \leq x \leq 0.5$) to Eqs. (2) and (4).

| | $x=0.1$ | | $x=0.25$ | | $x=0.5$ | |
|---|---|---|---|---|---|---|
| Shift | $K_0$ | $K^0_{anom}$ | $K_0$ | $K^0_{anom}$ | $K_0$ | $K^0_{anom}$ |
| $K_{\mu,1c}$ | 0.00087(7) | 0.00034(1) | 0.00063(2) | 0.00038(4) | 0.00062(4) | 0.00061(13) |
| $K'_{\mu,1c}$ | 0.0005(2) | −0.00024(8) | 0.00045(4) | −0.00031(6) | 0.00047(3) | 0.00050(13) |

behavior. In $Ce_2IrIn_8$ there are two such sources: CEF effects and $T^*$ effects, the former near $T_{CEF} \cong 50$ K and the latter near $T^* = 15-20$ K. In principle these two effects yield different temperature dependencies for $K_\mu(T)$ on the high- and low-temperature sides of the anomaly: CEF effects should yield a roughly linear-linear relation (above and below $T_{CEF}$) and $T^*$ effects a linear-logarithmic behavior (as discussed above). In practice, however, experimental uncertainties and the limited temperature ranges which can be measured above and below the anomalies mean that additional information is often helpful, if not necessary, to distinguish these effects. In the case of $Ce_2IrIn_8$, for example, we have analyzed measurements of the zero-field, magnetic specific heat in $Ce_2IrIn_8$ taken by Kim *et al.*[12] between $T=0.4-8$ K. These data show that the magnetic contribution to $C/T$ depends weakly on temperature between 5 and 8 K. Making the assumption that $C/T$ continues to be approximately temperature-independent between 8 K and 17 K, we find that $\cong R$ ln 2 of entropy is released between $T=0$ and $T \cong 17$ K. The release of $\cong R$ ln 2 of entropy between $T=0$ and $T=T^*$ is considered to be one signature of the Kondo-liquid state.[2] Thus, the magnetic specific-heat data corroborate our assignment of the low-temperature Knight-shift anomalies in $(Ce_{1-x}La_x)_2IrIn_8$ to $T^*$ effects.

As discussed in the introduction, a preliminary report of $Ce_2IrIn_8$ muon Knight-shift data taken only for $H=1$ T was presented earlier.[14] The data presented here in applied fields up to 3 T (where the shifts are better defined than in 1 T field), and the performance of an angular scan for the Knight shifts, have allowed us to make a definitive identification of three muon sites in $(Ce_{1-x}La_x)_2IrIn_8$. In contrast, only two muon sites were assumed in the preliminary analysis.[14] Furthermore, the additional data and a comparison with the magnetic specific heat have allowed us to cleanly separate the Knight-shift anomalies resulting from $T^*$ and $T_{CEF}$.

Our analysis yields a small, but statistically significant, difference in the estimations of $T^*$ for $H \parallel c$ and $H \parallel a$ axes in tetragonal $Ce_2IrIn_8$. This anisotropy could be due to systematic errors in the overall analysis on the order of 5 K (from uncertainties associated with subtraction of the $B\chi$ term in Eq. (3), for example), or may result from the effect of the applied field on a slightly anisotropic, intersite $f$-electron coupling. The latter would lead to the conclusion that $T_a^* \cong T_c^*$ for zero-applied field in tetragonal $Ce_2IrIn_8$. This is important because if $T^*$ is the characteristic temperature by which $\sim R$ ln 2 of entropy associated with the spin degrees of freedom of the KL is released, then there is no obvious reason why it should be strongly anisotropic in zero or small applied fields ($\gamma_{Ce}\hbar H \ll k_B T^*$). Nevertheless, we note that because our measurements were carried out in finite fields, we cannot strictly conclude that $T_a^* = T_c^*$ in zero field.

The result that $T_a^* \cong T_c^*$ in $Ce_2IrIn_8$ is similar to the analysis of NMR Knight-shift anomalies in $CeRhIn_5$ and $UPt_3$.[7] In contrast, highly anisotropic $T^*$ values ($T_c^* \cong 2-3T_a^*$) were obtained from NMR data in $CeCoIn_5$ and $CeCu_2Si_2$.[7] However, the effects of CEFs may not have been properly accounted for in the analysis of the latter materials. For example, inelastic neutron scattering reveals[17] two excited CEF levels in $CeCoIn_5$ at $T=283$ and 100 K, and the reported[7] $T^*$ values are $T_c^*=93$ K and $T_a^*=43$ K. In light of the fact that zero-field thermodynamic data in $CeCoIn_5$ yield $T^* \cong 45$ K,[6] it seems plausible that the Knight-shift anomaly near $T=100$ K attributed to $T_c^*$ in $CeCoIn_5$ is due to a depopulation of the first-excited CEF level instead.

The situation in $CeCu_2Si_2$ is less clear. The reported $T^*$ values for $CeCu_2Si_2$ are $T_c^*=171$ K and $T_a^*=58$ K (Ref. 7) based on NMR experiments by Ohama *et al.*, who attributed the broad Knight-shift anomaly in the region of 50–70 K to CEF effects.[4] There has been controversy in the assignment of CEF levels in $CeCu_2Si_2$, however. Early inelastic neutron experiments[18] found CEF levels at 140 and 364 K, but later neutron experiments[19] could confirm only the higher-lying level. It is also worth noting that this compound has a long history of metallurgical difficulties producing variations in competing low-temperature magnetic and superconducting states. Thus, the situation in $CeCu_2Si_2$ remains inconclusive. It is difficult to see how field-induced anisotropies of a factor of three in $T^*$ could be produced by the in-field NMR measurements, however.

To conclude, most Knight-shift data from $\mu$SR and NMR in noncubic materials are consistent with nearly isotropic $T^*$ values in relatively small applied fields, suggesting that $T^*$ may be isotropic in zero field. Corroborating in-field, magnetic specific-heat measurements would be useful in further clarifying this issue. Finally, we note that our data in $(Ce_{1-x}La_x)_2IrIn_8$ show that both the Ce-rich and Ce-depleted muon sites yield consistent values of $T^*$, indicating that $T^*$ is not a local quantity, but is a property of the overall $f$-electron lattice. The reduction of $T^*$ with increasing La concentration shows that the intersite interaction between $4f$ electrons becomes systematically weaker in the La-doped systems, affecting the onset of the coherent heavy-electron state, as seen previously in $Ce_{1-x}La_xCoIn_5$.[6]

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