

CeCu₂Si₂: More nearly magnetic than UBe₁₃

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We have prepared and studied, via dc and ac susceptibility and specific heat, samples of Ce_{1-x}M_xCu_{2.2}Si₂, with $M=Y, La,$ and Lu . All samples show an increase in the Wilson ratio, $\propto \chi/\gamma$, with the low-temperature χ per Ce mole increasing from Lu to Y to La and Ce_{1-x}La_xCu_{2.2}Si₂ actually having $\chi(1.8\text{ K})$ per Ce mole larger than in pure CeCu_{2.2}Si₂ for $x > 0.5$. The low-temperature specific heat of CeCu_{2.2}Si₂ is enhanced by dilution by La , when normalized per Ce mole, whereas dilution by Y and Lu strongly depresses the per-Ce-mole specific heat. These two contrasting effects may be linked to a volume effect, since Y and Lu contract the CeCu_{2.2}Si₂ lattice while La expands it. The fact that dilute Ce in the expanded host lattice Ce_{1-x}La_xCu_{2.2}Si₂ has increased χ and γ at low temperatures argues for a nearness to magnetism (substantiated by recent work on Ce_{1-x}Th_xCu_{2.2}Si₂) that contrasts with results for U_{1-x}M_xBe₁₃.

I. INTRODUCTION

With the discovery¹ of superconductivity in CeCu₂Si₂ a great deal of interest has been focused on $4f$ and $5f$ heavy-fermion systems. Some have been found to be superconducting (CeCu₂Si₂, UBe₁₃, and UPt₃) while others (e.g., U₂Zn₁₇ and UCd₁₁) display the magnetism expected for such highly correlated electrons, while still others (including CeAl₃ and CeCu₆) display no long-range order.² It has become evident with further study of these materials that the nonmagnetically ordered systems are more nearly magnetic than previously understood (with the exception of UPt₃, which was always³ known as a spin fluctuation system). For example, μ SR work⁴ on CeAl₃ has found magnetic correlations that are partly coherent (albeit spatially inhomogeneous and frustrated) below 0.7 K; doping experiments⁵ in CeCu₆ have shown that very little perturbation (CeCu_{5.91}Ag_{0.09}) causes long-range magnetic order of apparently antiferromagnetic nature.

The question of whether these ubiquitous magnetic correlations in heavy fermion systems are tied to the occurrence of superconductivity in CeCu₂Si₂, UBe₁₃, and UPt₃ is an often considered but as yet unsolved speculation. A second important, and as yet unresolved, question is the source of the heavy fermion ground state, i.e., what causes the observed huge effective electron masses.

Recently, significant progress has been made⁶ on this latter question in UBe₁₃ with the discovery that 40% of the large m^* is due to single-ion effects, and that the many-particle contribution to m^* is extremely sensitive to the U-Be separation. Also, no evidence for nearly magnetic behavior was found⁶ in any of the U_{1-x}M_xBe₁₃ alloys studied, for all nine of the M elements investigated ($M=Hf, Zr, Sc, Lu, Y, Pr, Ce, Th,$ and La) which form nonmagnetic MBe_{13} compounds. Thus, a similar study of Ce_{1-x}M_xCu₂Si₂ was undertaken and is reported here to answer the two questions: (1) How nearly magnetic is CeCu₂Si₂, as evidenced by the response to doping?; (2) is it possible to separate single-ion and many-particle effects

in Ce_{1-x}M_xCu₂Si₂ by studying the dilute Ce limit?

Despite the many studies on CeCu₂Si₂ since the discovery¹ of superconductivity therein in 1979, there are no published studies of the specific heat, C , of Ce_{1-x}M_xCu₂Si₂ for $x > 0.2$. (The effective mass m^* is proportional to $\gamma \equiv C/T$ as $T \rightarrow 0$.) Existing studies on C on Ce_{1-x}M_xCu₂Si₂ alloys are limited to $T \leq 1.5$ K, although numerous studies exist on resistivity (and for $M=La$, susceptibility) up to 300 K.

In any study of the low-temperature properties of CeCu₂Si₂, it is desirable to use excess Cu (e.g., Cu_{2.2}) to avoid spurious magnetic effects due to uncompensated Ce³⁺ ions, as have been well documented in the magnetic susceptibility^{2,7} [where $\chi(T \rightarrow 0)$ is enhanced at low field in Cu deficient samples, while superconductivity is suppressed] and the resistivity (where⁸ the low-temperature peak shifts downward in temperature and is enhanced in magnitude in Ce-deficient samples). In the specific heat, there is a peak around 3 K amounting to about one-third of the total C whose size and position in temperatures are mildly (± 1 K, $\pm 10\%$ in size) dependent^{2,9} on Cu stoichiometry. This peak is broad enough and small enough that it is not evident² in a plot of C/T (the monotonic upward trend in C/T as T is lowered is dominated by the change in T).

The choice of M , which must also of course form MCu_2Si_2 in the ThCr₂Si₂ structure as does CeCu₂Si₂ so as to provide an unlimited intersolubility of Ce and M in Ce_{1-x}M_xCu₂Si₂, is partly based on wanting a low vapor pressure so as to prepare the samples by arc melting. This argues against Ca, Sm, Dy, Er, Tm, Sr, Eu, and Yb. Also, it is desirable to avoid known magnetic MCu_2Si_2 compounds [$M=Gd(T_N=12\text{ K})$,¹⁰ $M=Np(T_c=41\text{ K})$,¹¹ $M=U(T_c=100\text{ K})$,¹² $M=Ho(T_N=8\text{ K})$]¹³ which would obscure any attempt to determine the nature of the magnetic correlations due to the Ce in Ce_{1-x}M_xCu₂Si₂. Also, magnetic transitions in C in MCu_2Si_2 would make difficult the determination of γ at low temperatures to determine the effective mass of the dilute Ce ions. These

two criteria leave $M=Y, La, Th, Lu, Pr,$ and Tb . We have recently published¹⁴ our findings for $M=Th$, where antiferromagnetism occurs for $x \geq 0.1$ (apparently¹⁴ coexistence with superconductivity up to $x=0.25$), with more ferromagneticlike behavior setting in for $x > 0.5$. In order to keep the present study focused on the issue of the magnetic and m^* behavior of the Ce $4f$ electrons, Pr and Tb have been left to later work. [$\chi(4.2\text{ K})$ for $PrCu_2Si_2$ is¹³ about 30 times that of $CeCu_2Si_2$, and $TbCu_2Si_2$ is thought¹⁵ to be metamagnetic.]

Thus, we have prepared samples of $Ce_{1-x}M_xCu_{2.2}Si_2$ for $M=Y, La,$ and Lu ($x=0, 0.1, 0.2, 0.5, 0.9,$ and 1.0) via arc melting, annealed them at 900°C for one week (which optimized¹⁶ the homogeneity of $CeCu_{2.2}Si_2$) and characterized them with specific heat (primarily for $1\text{ K} < T < 10\text{ K}$) and dc susceptibility ($1.8\text{ K} \leq T \leq 400\text{ K}$). Resistivity data exist in the literature for $Ce_{1-x}La_xCu_2Si_2$, $0 \leq x \leq 1$, and over a more limited range for $Ce_{1-x}Y_xCu_2Si_2$. These data will be referred to briefly below; the lack of Cu excess in these samples will not hamper the general inferences needed in the discussion of the present work.

II. RESULTS AND DISCUSSION

Lattice parameters and unit cell volumes for $MCu_{2.2}Si_2$, $M=Ce, Y, La,$ and Lu , are given in Table I. X-ray diffraction studies of all of the pseudoternary samples indicated single-phase material. The a and c lattice parameters are monotonic in each $Ce_{1-x}M_xCu_{2.2}Si_2$ system. In general, Y and Lu contract, while La expands the volume of the $CeCu_{2.2}Si_2$ lattice.

The dc magnetic susceptibility expressed per Ce mol and with (negligible) contributions for the pure $MCu_{2.2}Si_2$ subtracted, at 1.8 K for $Ce_{1-x}M_xCu_{2.2}Si_2$ is shown in Fig. 1, $M=Y, La,$ and Lu . [For our annealed sample of pure $CeCu_{2.2}Si_2$, $\chi(1.8\text{ K})=7.4 \times 10^{-3}$ emu/mol in good agreement with the value reported for $CeCu_{2.2}Si_2$ in Ref. 7.] The magnitude of the low-temperature susceptibility per Ce mol compared to that for pure $CeCu_{2.2}Si_2$ falls by 50% (70%) for 10% Ce in the (contracting) host lattice $Y(Lu)Cu_{2.2}Si_2$ [i.e., $Ce_{0.1}Y_{0.9}(Lu_{0.9})Cu_{2.2}Si_2$], while it increases 120% for 10% Ce in the (expanding) host lattice $LaCu_{2.2}Si_2$. Since Y and La are isoelectronic, as a first approximation this large difference in behavior between the two may be considered a volume effect. Since $Ce_{1-x}Lu_xCu_{2.2}Si_2$ is even more contracted than for $M=Y$ (see Table I), the even greater decrease in $\chi(1.8\text{ K})$

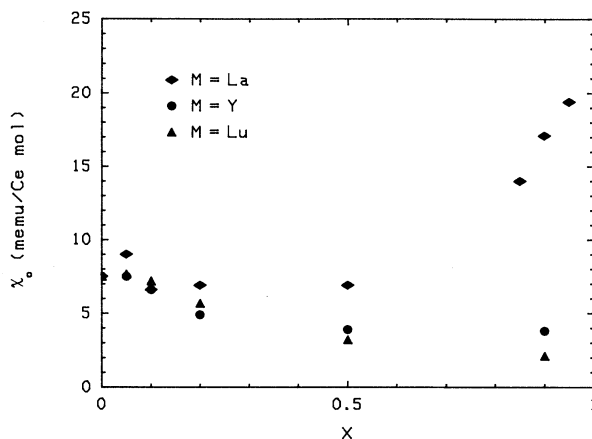


FIG. 1. Low temperature, $T=1.8\text{ K}$, magnetic susceptibility for $Ce_{1-x}M_xCu_{2.2}Si_2$, $M=Y, Lu,$ and La , as a function of x normalized per Ce mol via $[\chi(Ce_{1-x}M_xCu_{2.2}Si_2) - x * \chi(MCu_{2.2}Si_2)] / (1-x)$.

for Lu versus Y is consistent with this volume effect explanation. This marks the first difference in our present work with the results for $U_{1-x}M_xBe_{13}$, where $\chi(1.8\text{ K})$ per U mol was constant and independent of M for all nine M elements studied, again where MBe_{13} was nonmagnetic just as in the present work.

From plots of $1/\chi$ versus T, μ_{eff} , the high-temperature effective magnetic moment, may be obtained from the Curie-Weiss law behavior (i.e., $1/\chi$ is linear in T) above 200 K for these samples. [To obtain the values per Ce mol, the respective $\chi(MCu_{2.2}Si_2)$ multiplied by x is first subtracted and then the remainder is normalized by $1/(1-x)$.] These μ_{eff} values are shown in Table I; these values stay close to the $2.58\mu_B$ observed for pure $CeCu_{2.2}Si_2$ and do not track the variation in the low temperature $\chi(1.8\text{ K})$.

In order to better understand the behavior of $Ce_{1-x}M_xCu_{2.2}Si_2$, $M=Y$ and La , let us review the published resistivity data and then discuss the specific heat results of the present work.

There have been attempts¹⁷ in the literature to determine T_{Kondo} in $Ce_{1-x}La_xCu_2Si_2$ from the position of the low-temperature maximum in the resistivity (which varies² between 5 and 24 K even in undoped $CeCu_2Si_2$). Determining T_{Kondo} , the characteristic temperature of

TABLE I. Lattice parameters and μ_{eff} for $Ce_{1-x}M_xCu_{2.2}Si_2$. a for pure $CeCu_{2.2}Si_2$ is 4.105 \AA ; c for pure $CeCu_{2.2}Si_2$ is 9.933 \AA ; unit cell volume for pure $CeCu_{2.2}Si_2$ is 167.4 \AA^3 .

x	$a(\text{\AA})$	$c(\text{\AA})$	Unit cell volume (\AA^3)	$\mu_{\text{eff}}(\mu_{\text{Bohr}})$
	Y/La/Lu	Y/La/Lu		
0.05	4.088/4.094/4.085	9.914/9.900/9.916	165.6/165.9/165.5	2.8/2.58/2.68
0.10	4.083/4.100/4.084	9.930/9.911/9.905	165.5/166.6/165.2	2.59/2.58/2.65
0.20	4.070/4.107/4.059	9.923/9.909/9.915	164.4/167.1/163.3	2.69/2.65/2.66
0.50	4.027/4.118/3.999	9.929/9.924/9.952	161.0/168.3/159.2	2.75/2.66/2.82
0.90	3.973/4.143/3.930	9.972/9.923/9.966	157.4/170.3/153.9	2.84/2.57/2.85
1.0	3.967/4.149/3.908	9.959/9.918/9.957	156.7/170.7/152.1	

the process whereby the conduction electrons compensate the local magnetic moment, from the peak in the low-temperature resistivity has been shown^{8,18} to give fallacious results. A preferable way is to use $\gamma \propto 1/T_K$, with the specific heat γ defined as $C/T(T \rightarrow 0)$. In any case, resistivity data of $Ce_{1-x}La_xCu_2Si_2$ has been reported.^{8,17,19} The peak in ρ at 20 K for $x=0$ moves to lower temperature with increasing x , $\rho(300\text{ K})$ decreases monotonically with increasing x , and $\rho(T \rightarrow 0)$ rises with increasing x to a maximum around $x=0.5$, before falling again. The resistivity for $Ce_{0.97}M_{0.03}Cu_{2.2}Si_2$, $M=La$ and Y , has been reported.²⁰ The maximum in ρ is depressed in temperature more rapidly by Y than La , with the Y -doped sample having a higher $\rho(T \rightarrow 0)$ and the same $\rho(300\text{ K})$ as the La -doped sample. Other than the maximum in ρ in $Ce_{1-x}La_xCu_2Si_2$ occurring for $x=0.5$ (which is just caused by the maximal disorder scattering) corresponding to the minimum in $\chi(1.8\text{ K})$ versus x (Fig. 1), no correlations between the χ and ρ behavior seems compelling.

On the other hand, the specific heat γ values per Ce mol, see Figs. 2 and 3, for $Ce_{1-x}M_xCu_{2.2}Si_2$ ($M=Y, Lu$, and La) show a rather strong correlation with the $\chi(1.8\text{ K})$ versus x behavior. The γ value per Ce mol for $Ce_{1-x}M_xCu_{2.2}Si_2$, $M=Y$ and Lu , falls by over a factor of 6 between pure $CeCu_{2.2}Si_2$ and $Ce_{0.1}M_{0.9}Cu_{2.2}Si_2$. Since m^* , the electron effective mass, depends² monotonically on γ , this implies a sharp decrease in the Ce 4*f*-heavy electron effective mass with increasing x in $Ce_{1-x}M_xCu_{2.2}Si_2$, $M=Y$ or Lu , i.e., that dilute Ce in a

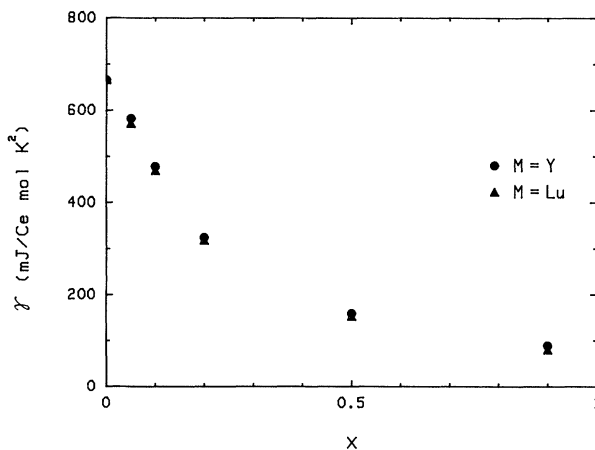


FIG. 2. Shown are C/T values at 1.1 K for $Ce_{1-x}Y_x(Lu)Cu_{2.2}Si_2$ as a function of x , calculated as $[C/T(Ce_{1-x}M_xCu_{2.2}Si_2) - C/T(MCu_{2.2}Si_2)]/(1-x)$ at 1.1 K to achieve a per Ce mol normalization. The full value (instead of x^* as for χ) of $Y(Lu)Cu_{2.2}Si_2$ is subtracted to approximate the contribution of the Ce 4*f* electrons, with the lattice contribution fully subtracted. [γ for $Y(Lu)Cu_{2.2}Si_2$ is only 3.2(3.0) mJ/mol K^2 and $\Theta_D=400(379)$ K.] Since, as will be seen in a later figure, C/T is not rapidly varying with temperature at 1 K, these 1.1-K values may be used to approximate $\gamma(\equiv C/T$ as $T \rightarrow 0)$, which is proportional to m^* . Thus, upon dilution with Y and Lu , Ce 4*f* electrons in the $CeCu_2Si_2$ lattice have a monotonic decrease in m^* .

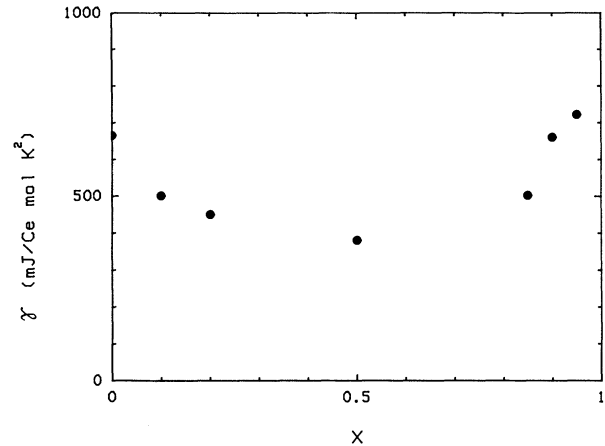


FIG. 3. C/T (1.1 K) for $Ce_{1-x}La_xCu_{2.2}Si_2$ normalized as in Fig. 2. The $x=0.9$ sample measured to 0.3 K showed no evidence of a magnetic transition. Thus, upon dilution with La , Ce 4*f* electrons in the $CeCu_2Si_2$ host lattice experience, after an initial decrease, an enhancement of m^* with further decrease in Ce concentration in sharp contrast to the results for isoelectronic Y and for Lu .

$Y(Lu)Cu_{2.2}Si_2$ host lattice, with its $8\frac{1}{2}\%$ (9.2%) smaller unit cell volume¹⁶ does not have single-ion generated large m^* . Thus, closer Ce-Cu distances are not conducive to high m^* for the Ce 4*f* electrons. This is unlike the case of $U_{1-x}M_xBe_{13}$, where as doping by smaller M ions decreased the U-Be distance, γ per U mol decreased by 50% by $x=0.2$, but then γ per U mol remained constant with further increase in x , independent of further d_{U-Be} decrease.

The values of the specific heat per Ce mol for $Ce_{1-x}La_xCu_{2.2}Si_2$, Fig. 3, show the same (although not as rapid) initial all-off of γ with increasing x as seen for Y . However, as Ce is further diluted by La , the low-temperature specific heat γ per Ce mol increases until at

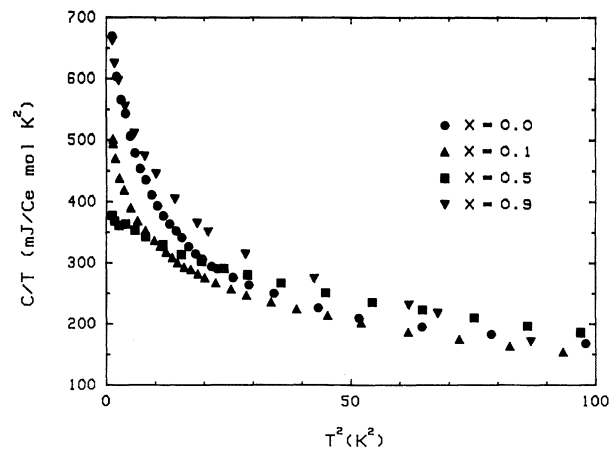


FIG. 4. Specific heat divided by temperature vs temperature squared for $Ce_{1-x}La_xCu_{2.2}Si_2$ and normalized per Ce mol as for Fig. 2. Note that the rise in C/T below 10 K is preserved for all x , and even slightly enhanced for $x=0.9$.

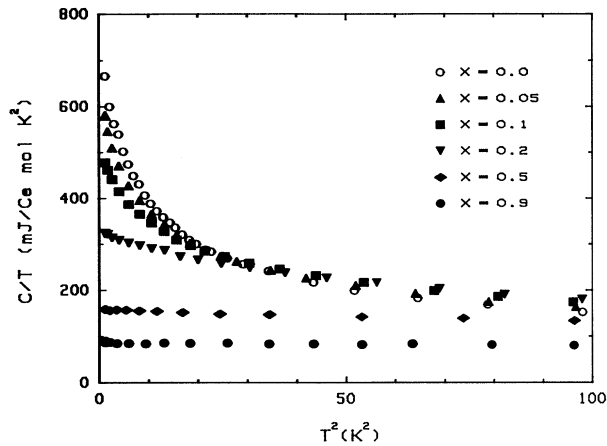


FIG. 5. Low-temperature specific heat of $Ce_{1-x}Y_xCu_{2.2}Si_2$ divided by temperature vs temperature squared with C/T of pure $YCu_{2.2}Si_2$ subtracted and normalized by $1-x$ as for Fig. 3. Clearly, the formation of the heavy fermion ground state below 10 K (i.e., the increase in C/T as $T \rightarrow 0$, with $m^* \propto \gamma$) is severely impeded already by 20% Y, and is essentially halted by 50% Y.

$Ce_{0.1}La_{0.9}Cu_{2.2}Si_2$ the γ per Ce mol of pure $CeCu_{2.2}Si_2$ is attained. In order to investigate this behavior, the specific heat of the $x=0.9$ sample was measured to lower temperature (0.3 K) and a further composition, $x=0.95$, was prepared and characterized.

The specific heat of $Ce_{0.1}La_{0.9}Cu_{2.2}Si_2$ showed no sign of magnetic ordering down to 0.3 K. Thus, even though the $\chi(1.8 \text{ K})/\text{Ce mol}$ is enhanced by a factor of 2 for this sample, the increase in the low-temperature γ value is evidently not due to a magnetic ordering anomaly causing an upturn in C/T . In fact, C/T rises more steeply below 5 K for pure $CeCu_{2.2}Si_2$ than for $Ce_{0.1}La_{0.9}Cu_{2.2}Si_2$, as seen in Fig. 4.

The $\gamma(1 \text{ K})$ per Ce mol value for the $Ce_{0.05}La_{0.95}Cu_{2.2}Si_2$, shown in Fig. 3 [the $\chi(1.8 \text{ K})$ value is shown in Fig. 1] continues the upward trend in γ per Ce mol with decreasing Ce concentration. This increase in γ per Ce mol upon dilution for $x > 0.5$ is a surprising difference from the behavior of the isoelectronic $Ce_{1-x}Y_xCu_{2.2}Si_2$ and a total departure from our previous doping results⁶ in $U_{1-x}M_xBe_{13}$. The specific heat between 1 and 10 K [normalized by subtracting of C of $MCu_{2.2}Si_2$ and division of this difference by $(1-x)$ to achieve the per Ce mol result], Figs. 4, 5, and 6 further emphasize the difference between $LaCu_{2.2}Si_2$ and $Y(Lu)Cu_{2.2}Si_2$ as host lattices for Ce. Above 5 K, the specific heat for $M=La$ is essentially independent of x (see Fig. 4), whereas the specific heat over the whole 1–10-K regime depends on x for $x > 0.2$ for $M=Y(Lu)$ (Figs. 5 and 6). Interestingly, almost the same specific

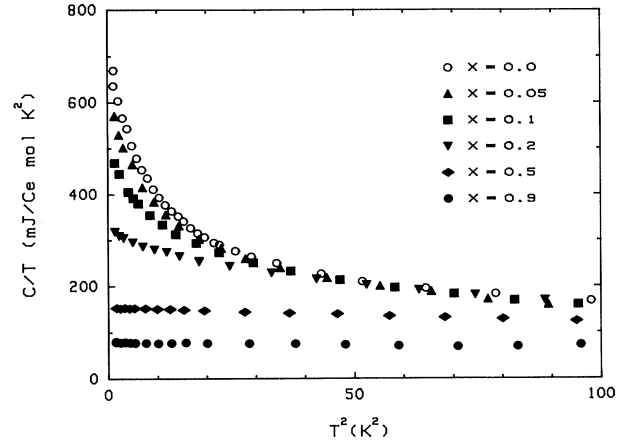


FIG. 6. Low-temperature specific heat of $Ce_{1-x}Lu_xCu_{2.2}Si_2$ divided by temperature vs temperature squared with C/T of pure $LuCu_{2.2}Si_2$ subtracted and normalized by $1-x$ as for Figs. 2–4. These results for Lu are remarkably similar to those presented in Fig. 5 for Y.

heat behavior as seen in $Ce_{1-x}La_xCu_{2.2}Si_2$ is observed^{21,22} in $Ce_{1-x}La_xCu_6$, where above 2 K, C/T per Ce mol is²² composition independent, while below 1 K, C/T per Ce mol is higher (by as much as 50%) for $x=0.9$ compared to pure $CeCu_6$.

What the exact underlying reasons for this increase in $\gamma/\text{Ce mol}$ upon dilution in $Ce_{1-x}La_xCu_{2.2}Si_2$ are not clear. The accompanying increase in the low-temperature magnetic susceptibility is suggestive, however, that magnetic correlations between the dilute Ce 4f ions mediated by the conduction electrons in the expanded lattice play a role.

III. SUMMARY

By preparing and studying $Ce_{1-x}M_xCu_{2.2}Si_2$, $M=Y, Lu$ and La , we have found sharp changes in magnetic susceptibility and in the effective mass ($\alpha \gamma$) of the Ce 4f electrons as a function of doping. Although isoelectronic, Y and La show contrasting results, with m^* and low temperature χ monotonically decreasing with increasing x for $M=Y$ (as well as for Lu) while for $M=La$ m^* and $\chi(1.8 \text{ K})$ go through a minimum and rise to values per Ce mol larger than for pure $CeCu_{2.2}Si_2$ with increasing x . These differences are likely volume effects, with Y and Lu contracting and La expanding the $CeCu_{2.2}Si_2$ lattice.

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