Title
Characterization of single crystals of CeCu2Si2. A source of new perspectives

Journal
Physical Review B, 28(1)

ISSN
2469-9950

Authors
Stewart, GR
Fisk, Z
Willis, JO

Publication Date
1983-07-01

DOI
10.1103/physrevb.28.172

Characterization of single crystals of $CeCu_2Si_2$. A source of new perspectives

G. R. Stewart, Z. Fisk, and J. O. Willis
Los Alamos National Laboratory, Los Alamos, New Mexico 87545
(Received 17 January 1983; revised manuscript received 28 March 1983)

We report the first thorough characterization of single crystals of $CeCu_2Si_2$. Measurements on these flux-grown crystals, which are not superconducting above 0.050 K, include ac susceptibility, resistivity, Hall effect, and specific heat. A review of other measurements is given, and the implications of our single-crystal data are discussed. Specifically, our data are consistent with superconductivity in $CeCu_2Si_2$ being destroyed by having too low a Kondo temperature, although T_{Kondo} is not found to be inversely proportional to γ as previously claimed. The entropy associated with the low-temperature specific-heat anomaly is found to be only $0.66R$ ln2, in contrast to the previous result of R ln2 for polycrystalline material. The lack of superconductivity in our single crystals does not appear to be due to poor stoichiometry to ±5%, as proposed previously for single crystals grown by a Bridgman technique. The possibility of charge-density waves suppressing superconductivity in strain-free material is discussed, although no experimental verification is found.

I. INTRODUCTION

One of the more fascinating superconducting materials to be discovered in the last several years is $CeCu_2Si_2$. Steglich *et al.*[1] reported that they had found a bulk specific-heat anomaly ΔC at about 0.5 K in three different unannealed polycrystalline samples of $CeCu_2Si_2$, which they associated with resistive and inductive indications of superconductivity at slightly higher temperatures. This discovery aroused great interest because of the presence of an enormous γ (1 J/mole K^2) in these low-temperature specific-heat (LTSH) data, as well as an enormous ΔC, i.e., $\Delta C/\gamma T_c \sim 0.85$. This indicated that electrons with strong electron-electron correlations were taking part in the superconductivity. To place this in perspective, the other material's system, which is today considered[2] an example of a "heavy fermion" superconductor, U_6Fe, has a γ which, when normalized per U atom (presumably the Fe in U_6Fe and the Cu and Si in $CeCu_2Si_2$ make little contribution to γ), is only about $\frac{1}{40}$ of that for $CeCu_2Si_2$.

Since Steglich *et al.*[1] first announced their surprising result, a number of further measurements[3–10] have been performed on $CeCu_2Si^2$ in order to better understand this system. Unfortunately, numerous puzzles remain, including the existence of significant sample dependence of measured properties, including $\Delta C/\gamma T_c$, upper critical field,[5] and even T_c.[6,7] In fact, Hull *et al.*[6] observe no T_c resistively down to 0.060 K in a polycrystalline sample prepared similarly[6] to those of Steglich *et al.* The Meissner effect has been used to characterize many of the $CeCu_2Si_2$ samples studied and varies[3] from less than 1% in some unpowdered polycrystalline specimens to 60% in one powdered specimen, where powdering is thought[3] to allow better flux penetration. In addition to the sample dependence of $\Delta C/\gamma T_c$, Hc_2, T_c, and dc magnetization, γ varies from approximately 1 to 1.3 J/mole K^2, depending on the sample.

With the exception of some resistivity as a function of pressure data on a single crystal of $CeCu_2Si_2$ taken by Aliev *et al.*,[9] and T_c measurements on Cu-deficient single crystals by Bredl *et al.*,[7] all the characterization to date on $CeCu_2Si_2$ has been on polycrystalline samples. Since sample quality plays such a crucial role in determining the properties of $CeCu_2Si_2$, it is important to closely compare materials prepared by different methods. We have grown single crystals of $CeCu_2Si_2$ from flux and have characterized them by x-ray diffraction, x-ray fluorescence, energy dispersive spectrometry, ac susceptibility (0.050—4 K), resistivity (1.4—300 K; 0—11 T), Hall effect (2—160 K), and specific heat (0.3—33 K, 0—10 T).

II. EXPERIMENTAL

Single-crystal plates of $CeCu_2Si_2$, typically 0.2—0.3 mm thick and up to 5 mm on a side, were grown from a liquid-In flux. The particular crystals used in these measurements were grown by slow cooling an In:$CeSi_2$:Cu mixture (0.95:0.01:0.04 atomically) in an alumina crucible from 1400 °C to 500 °C at 4°/h. The amount of $CeCu_2Si_2$ relative to In can be greatly increased and a somewhat lower starting temperature is possible. The crystals were leached from the In matrix with HCl.

Since the Hall effect has not been performed as a function of temperature on polycrystalline $CeCu_2Si_2$, we also prepared such material by arc melting high-purity starting materials in an argon arc furnace. Weight losses of up to 0.5 wt. % were observed in these melts of 5 g typical size. The nature of this boil off was not determined.

The superconducting transition temperature T_c of both the single-crystal and polycrystalline material was measured using ac susceptibility measurements between 0.050 and 4.0 K. The measured single crystal shows a very slight anomaly at 0.5 K that was within the limits of error (i.e., less than approximately 1% of the sample was superconducting), whereas the polycrystalline material showed a transition into the superconducting state at 0.5 K.

This result is similar to that of Bredl *et al.*,[7] who removed a crystallite of dimension $2\times2\times0.1$ mm^3 from a 40-g boule and found no superconductivity down to 0.020 K. Bredl *et al.*[7] state that this lack of superconductivity in their single crystal is perhaps due to the 20% Cu deficiency measured for their crystallite. In order to test this

proposition for our (quite differently prepared) single crystals, we have done both x-ray fluorescence and energy dispersive spectroscopy measurements, using $CeCu_2$ as a standard. The energy dispersive spectroscopy indicated a 4.3% smaller Ce-to-Cu ratio in the standard than in the single crystal. However, the x-ray fluorescence measurement indicated a 4—5.5% greater Ce-to-Cu ratio in the standard than in the single crystal. Therefore, within our experimental error of ±5%, the Ce-to-Cu atomic ratio in our single crystals is 1:2. This still allows the possibility that the single crystals have the wrong Ce (and Cu) ratio to Si. We used a Gandolfi attachment to a Debye-Scherrer powder camera to determine the lattice parameter of a single crystal of $CeCu_2Si_2$, which occurs in the tetragonal $ThCr_2Si_2$ structure. The resultant lattice parameters ($a=4.101\pm0.001$ Å; $c=9.936\pm0.003$ Å) are in good agreement with those reported[11] for polycrystalline $CeCu_2Si_2$ ($a=4.105$ Å; $c=9.933$ Å) and measured by us on our polycrystalline material ($a=4.099$ Å; $c=9.924$ Å). Therefore, we conclude that the stoichiometry in our $CeCu_2Si_2$ crystals is comparable (to ±5%) to the correct 1:2:2 ratio.

Additionally, since the crystals were grown in an In flux, the question of In inclusions arises. The ac susceptibility measurements indicate the presence of In in the single crystals (i.e., an inductive anomaly at 3.4 K). Energy dispersive spectroscopy measurements place the amount of In as less than 0.5 at.%.

The resistance of a single-crystal plate was measured from 300 to 1.2 K using a standard four-probe ac technique at 220 Hz. The Hall effect on both a single crystal and a 0.4-mm slab of polycrystalline $CeCu_2Si_2$ was measured using a five-point dc method. In order to minimize contact resistance and therefore sample heating, 0.002-in. Pt leads were spot welded onto the samples. At temperatures below 10 K, a current of 5 mA was used. Above 10 K, 50 mA could be used without excessive sample heating. Hall voltage, V_H, was zeroed at zero field for both current directions at each temperature, and then fields of 5.5 and 11.0 T were applied. Within measurement error (10%), V_H went linearly with H and was independent in magnitude (but not sign) of current and field direction. Magnetoresistance measurements were made on both the single crystal and polycrystalline samples at the same time as the Hall-effect measurements.

The calorimeter used for measurements from 1.2 to 33 K and $H=0$ and 10 T has been described elsewhere.[12–14] A new sample platform of similar design but using an unencapsulated Cr 250 Ge thermometer from Cryocal, Inc. was used from 4 to 0.3 K, giving the specific heat of a vacuum-annealed 350-mg piece of 99.9999% pure Cu to within ±3% of the known[15] values.

Three single crystals of $CeCu_2Si_2$ with a total weight of 4.21 mg were then measured on this platform from 0.3 to 1.2 K in zero field and on our usual higher-temperature platform from 1.2 to 11 K in 0- and 10-T applied fields. Since the addenda correction to the total measured specific heat, which was $<0.5\%$ for $T<1.2$ K, grew to be too large a percentage (50%) by 7 K, a third measurement was made on a collection of ten single crystals (different from the first three measured), with a total weight of 15.56 mg, from 1.2 to 33 K in zero field. The addenda correction for this third measurement remained below 50% for all temperatures of measurement. The accuracy of all these specific-heat measurements is ±3%, except for the second run on 4.21 mg of material above 7 K, where the accuracy is only ±5%.

III. RESULTS AND DISCUSSION

A. Do charge-density waves exist in $CeCu_2Si_2$?

In addition to the unusual polycrystalline sample-dependent properties discussed in the Introduction, Aliev *et al.*[9] discovered that as pressure was applied up to 2.5 kbar, their single crystal (preparation method unstated), which had $T_c<0.05$ K at zero pressure, had its T_c increase monotonically to 0.5 K. Another system that immediately comes to mind[16] which has similar sample-dependent superconducting properties [$T_c\sim0.2$ K for single crystals,[17] 1 K (not bulk) for some polycrystals,[18] and 2 K at 10 kbar (Ref. 19)] is U. U has long been known to have an anomaly at 43 K in the specific heat, as well in other properties.[20] Recently, this anomaly has been shown[21] to be due to a charge-density wave. The existence of this charge-density wave (CDW) in U, which suppresses the superconductivity, is severely dependent on pressure as noted above, with only 10 kbar completely suppressing the CDW and allowing bulk superconductivity as measured by specific heat.[22] The polycrystalline material, on the other hand, has[22] no bulk superconductivity down to 0.1 K at zero pressure but has small regions that superconduct as high as 1 K due to a local suppression of the charge-density wave.

Is there a superconductivity-destroying charge-density wave at some temperature in single-crystal $CeCu_2Si_2$ which is suppressed by small applied pressures? Is this charge-density wave partially suppressed depending on details[4] of sample preparation in polycrystalline samples, where a large anisotropic thermal expansion is expected[7,9] to produce internal stresses?

In order to investigate whether a charge-density wave exists in single-crystal $CeCu_2Si_2$, we have measured the Hall effect in both single-crystal and polycrystalline material. The changes in Fermi-surface topology associated with the formation of a charge-density wave cause a sign change [in both U (Ref. 23) and the layered transition-metal dichalcogenide compounds[24]] in the Hall coefficient R_H,

$$R_H=V_Ht/BI\ , \tag{1}$$

where T is the thickness, B is the field transverse to the direction of the current I, and V_H is the voltage transverse to the direction of both the current and the field. It should be pointed out that the lack of a sign reversal in R_H does not rule out a charge-density wave in a given material, since the way in which the charge-density wave affects R_H will depend upon the details of the Fermi-surface topology.

The Hall voltage at 11 T and normalized to 50 mA of current is shown in Fig. 1 for a single crystal of $CeCu_2Si_2$, $t=0.14$—0.19 mm, and a polycrystalline sample, $t=0.4$ mm. There is no sign reversal for either sample out to the highest temperature of measurement. The ratio of V_H for the two samples is, within the limits of error, simply the inverse of the respective thicknesses, Eq. (1). Thus our

measurement of the Hall effect in $CeCu_2Si_2$ shows no sign of a charge-density wave in this material up to 160 K.

The Hall coefficient R_H corresponds to an electron carrier density n (using the simple formula $R_H = 4\times10^{-11}$ V cm/G A $= 1/ne$, where e is the electric charge) for both samples of 1.4×10^{21} cm^3 at 4.2 K. Aliev *et al.*[10] state n at 4.2 K for their polycrystalline $CeCu_2Si_2$ to be 2×10^{21} cm.^{3}. The Hall coefficient R_H found here is large compared to that of a simple metal ($R_H = 0.8\times10^{-11}$ V cm/G A for Cu) and is comparable to values found for the anomalous Hall effect in rare-earth metals.[25]

B. Resistivity

Our resistivity data on single-crystal and polycrystalline $CeCu_2Si_2$ is shown in Fig. 2. There has been a large range of resistivity behavior reported for $CeCu_2Si_2$.[26] The flat portion at 100 K in the resistivity is associated[26] with the crystal-field-split doublet and with the associated Kondo effect of Ce^{3+} which lies about 140 K above the ground-state doublet. The lower-temperature, $T_{\rho,0}$, peak in the resistivity is thought[7] to be due to Kondo-effect scattering, with $T_{\text{Kondo}} \sim \frac{1}{2}T_{\rho,0}$, off the ground-state doublet. Two different groups[7,9] have used the value of T_{Kondo} to explain the absence of superconductivity in certain samples of $CeCu_2Si_2$. The idea[7,9] that T_{Kondo} ($\propto T_{\rho,0}$) must be above a certain value for superconductivity to occur appears consistent with the $T_{\rho,0} \sim 6.5$ K value for our single crystals and the resistivity data under pressure of Aliev *et al.*[9] ($T_{\rho,0} = 6.3$ K, $dT_{\rho,0}/dP \sim 0.30$ K/kbar, and $dT_c/dP \sim 0.2$ K/kbar for $P \lesssim 2.5$ kbar in a single crystal). It means, however, that T_c is an extremely critical function of the exact value of $T_{\rho,0}$ for 6.3 K $\lesssim T_{\rho,0} \lesssim 7$ K. The other approach to connecting T_{Kondo} and T_c, via specific-heat data,[7] will be discussed for our single-crystal results below.

Resistivity data for both samples in an applied field of 11.0 T (not shown in Fig. 2) showed a negative magnetoresistance effect (characteristic of dense Kondo systems) of about 4.5% at 2 K. This change in resistance with field went to zero by 30 K for the polycrystalline sample but remained nonzero until 90 K for the single crystal (e.g., 2.1% decrease at 30 K and 11 T). Aliev *et al.*[10] found a negative magnetoresistance for polycrystalline $CeCu_2Si_2$ of 1.5% in a 4-T field below 40 K, which is inconsistent with our result both in magnitude and temperature dependence.

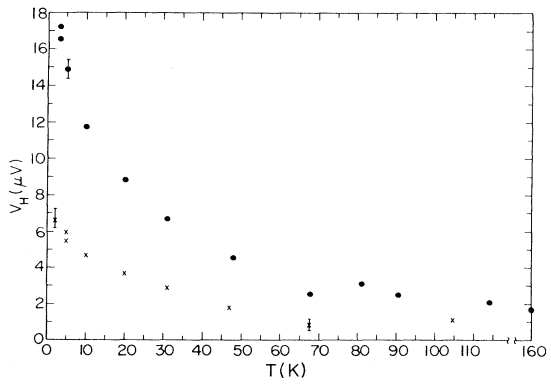

FIG. 1. Hall voltage V_H vs temperature for single-crystal (dots) and polycrystalline ($\times$'s) $CeCu_2Si_2$. When the Hall voltage fell below 2 μV, not only was the dc current reversed, but also the direction of the field was reversed to assure that a sign reversal of V_H was not obscured by noise.

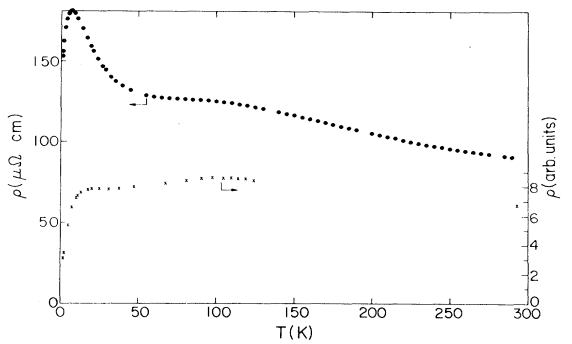

FIG. 2. Resistivity of single-crystal (upper curve, dots) and polycrystalline (lower curve, $\times$'s) $CeCu_2Si_2$.

C. Specific heat

The data are shown in Fig. 3 plotted as C vs $\ln T$, along with the data from Steglich *et al.*[1] It should be stressed that their data, which extend only to 7 K, and our data coincide, as best as we can determine from their published graph, to $\pm3\%$ below 2 K until their polycrystalline sample goes superconducting at ~0.5 K.

The temperature of the peak, $T_{\max}$, in our specific-heat data can be compared to T_{Kondo}; if we assume $T_{\text{Kondo}} \sim \frac{1}{2}T_{\rho,o} \simeq 3.5$ K, then $T_{\max}/T_{\text{Kondo}} \sim 0.6$, much different from the value of $\frac{1}{3}$ quoted for the data of Steglich *et al.*,[1] which was comparable to the dilute Kondo case.[4] The peak in our specific-heat data is lower in temperature than that for the Steglich *et al.*[1] data, and therefore T_{Kondo}(single crystal) $< T_{\text{Kondo}}$(polycrystalline) is again consistent with the argument[7,9] that T_{Kondo} must be above a certain temperature so that uncompensated Ce^{3+} $4f$ spins do not prevent superconductivity.

Two other comparisons of our specific-heat data to those of Steglich *et al.*[1] are informative. In order to facilitate these comparisons, our data are replotted in Fig. 4, with just the low-temperature data, as C/T vs T^2, and in Fig. 5 a similar plot is shown for all the data up to 33 K. In Fig. 4, if we describe C/T as equal to an electronic term γ and a lattice term βT^2, where the lattice term is $< 1\%$ of the electronic term, we see that γ increases 10% with just a 1 K temperature decrease. Such a rapid change of an electronic term, if such it is, with temperature is unusual. Such a negative slope of γ has also been noted above T_c in polycrystalline $CeCu_2Si_2$ by Lieke *et al.*[4] Although this behavior in γ vs T is at present unexplained, the value of $\gamma(T=0) = 1.05$ J/mole K^2 casts doubt on one of the important systematic behaviors reported by Steglich *et al.*[3] and by Bredl *et al.*,[7] i.e., their claim that $T_c = \text{const}\times\gamma^{-1}$ based on four samples. Our value of $\gamma = 1.05$ J/mole K^2 for γ is comparable with that obtained[3,7] for their highest T_c (0.6 K) material (as is the entire specific-heat behavior from 0.5 to 2 K, Fig. 3), and

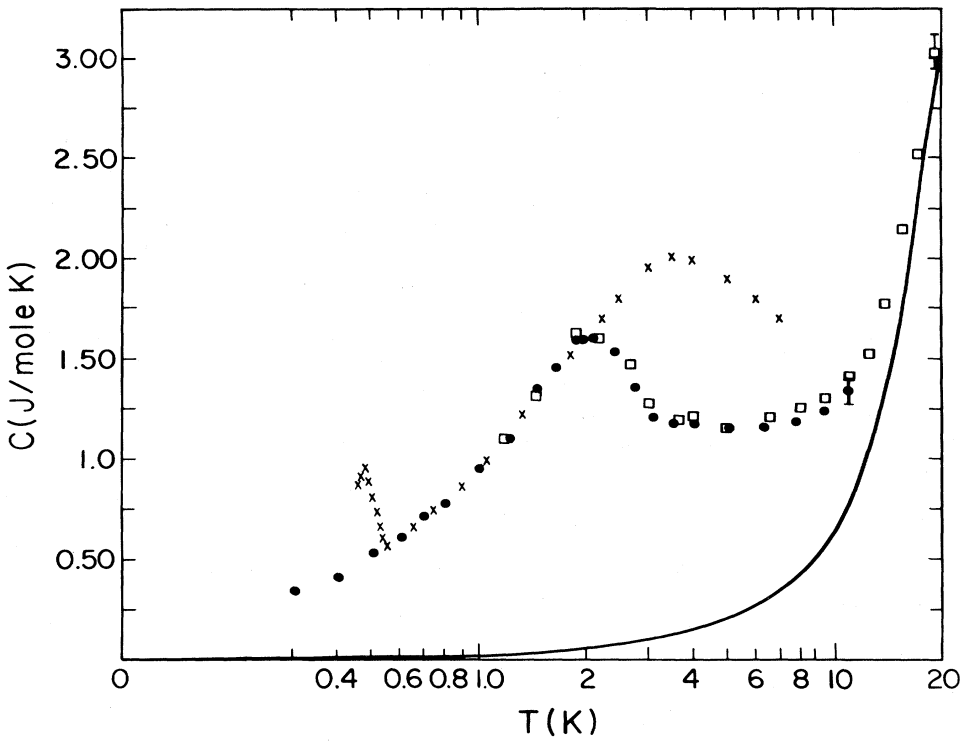

FIG. 3. Specific heat of single-crystal $CeCu_2Si_2$ from 0.3 to 19 K consisting of two measurements of differing sample masses (4.21 mg, dots; 15.56 mg, squares) as discussed in the text. The data of Steglich *et al.* (Ref. 1) shown as the $\times$'s were taken from their published graph and is only approximate. (It should be noted that only a small fraction of the Steglich *et al.* data is shown to preserve clarity.) Note the excellent agreement between the single-crystal and polycrystalline data between 0.6 and 2 K. The solid line drawn is an extrapolation of the higher-temperature data (see Fig. 5) following the relation $C=\gamma T+\beta T^3$ and represents the "normal" electronic and lattice contribution to the specific heat. Within the error limits of both sets of single-crystal data ($\pm 3\%$ for $T<7$ K), there is no sample dependence of the specific heat nor any aging effects over a period of two and one-half months.

yet T_c of our single crystals is <0.050 K. Also, these data show that one may not use $T_{\text{Kondo}}\sim 1/\gamma$, as was done[3,7] in the previous specific-heat work.

A second comparison between our specific-heat data and previous data which is of interest is the entropy S, associated with the low-temperature anomaly, which has the shape of a broadened Schottky anomaly. In order to determine the "normal" electronic and lattice contributions, Fig. 5 shows the data over the whole temperature range. The high-temperature $C/T=\gamma+\beta T^2$ behavior, when $\gamma=30$ mJ/mole K^2 and $\beta=0.3364$ mJ/mole K^4 above 19 K, is then extrapolated to lower temperatures

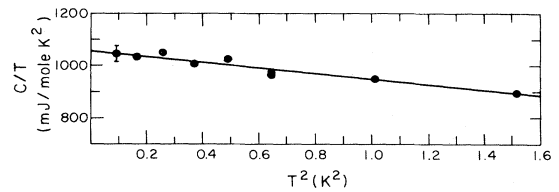

FIG. 4. C/T vs T^2 at low temperatures for single-crystal $CeCu_2Si_2$. This negative slope of γ with T has also been observed above T_c in polycrystalline material (Ref. 4).

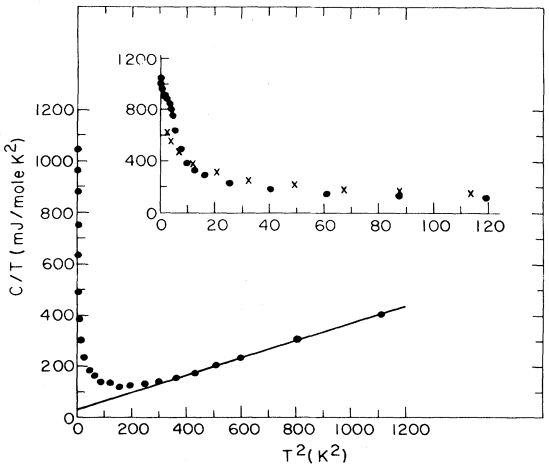

FIG. 5. Single-crystal data over the whole temperature range as well as data in 10-T applied fields ($\times$'s in the inset) taken on the same three crystals which were measured between 0.3 and 11 K. The straight line drawn through the higher-temperature data is used to calculate the entropy between 0.3 and 19 K ($=1.3$ J/mole K) that needs to be subtracted as due to the regular electronic and lattice behavior as discussed in the text.

(solid line, Fig. 3) and subtracted from the measured low-temperature specific heat. An entropy integral from 0 to 19 K of $(C/T)dT$ is then performed, giving $S=3.8$ J/mole K, which is 0.66 of the expected amount of entropy due to completely lifting the degeneracy of the low-lying doublet, i.e., R ln2 or 5.76 J/mole K. This is not in disagreement with work in dilute systems, where in at least one system only one-half of the expected entropy was measured.[27] However, Lieke *et al.*,[4] presumably referring to the data of Steglich *et al.*[1] shown in Fig. 3, claim that the anomaly in polycrystalline $CeCu_2Si_2$ has "an entropy connected with it of about $k_b$ln2 per Ce ion"[4] or R ln2 per Ce mole. Since, as discussed above in the experimental section, we are confident to $\pm 5\%$ of having the correct stoichiometry (and therefore Ce content) in our samples, we find this difference confusing. Partly, Lieke *et al.*[4] appear to have underestimated γ for "normal" $CeCu_2Si_2$ by using γ for $LaCu_2Si_2$ (stated[28] to be 4.4 mJ/mole K^2). Also, since the data of Steglich *et al.* extends only to 7 K, an extrapolation must have been made to higher temperatures and this extrapolation may have been in error.[29] Using a reasonable extrapolation of the Steglich *et al.*[2] data in Fig. 3 (a straight line through the three highest-temperature points shown in Fig. 3 and joining our data at 11 K), we find the entropy associated with the anomaly in polycrystalline $CeCu_2Si_2$ to be only 4.8 J/mole K—still well short of R ln2. It is interesting to note that Bloomfield and Hamann[30] predict the entropy under a spin-$\frac{1}{2}$ Kondo anomaly to be $0.45k_B$ per impurity in the dilute case, or just 0.65 of $k_B$ln2—to be compared to the $0.66R$ ln2 for a dense Kondo system reported here.

IV. CONCLUSIONS

We have seen one confirmation of previous work by our single-crystal characterization. Both our resistivity and specific-heat data on nonsuperconducting single-crystal $CeCu_2Si_2$ point to a low T_{Kondo} for this material, which is consistent with the earlier work[7,9] which found that T_{Kondo} had to be above a certain minimum value for superconductivity to occur.

We have measured the Hall effect of both single-crystal and polycrystalline material to investigate the possibility that there exist charge-density waves in $CeCu_2Si_2$ which might explain some of the unusual properties in this system, in analogy to U. No evidence for such a phenomenon was found, either in the temperature dependence of the Hall effect or in a room-temperature electron-diffraction experiment performed here by Rohr.

We have discovered three cases where propositions in the literature appear to be false: (1) T_{Kondo}, and therefore T_c, is not proportional to the apparent γ^{-1}, (2) the entropy under the anomaly of both polycrystalline and single-crystal material appears to be significantly less than R ln2 per mole of $CeCu_2Si_2$, (3) the lack of a superconducting transition in our single crystals of $CeCu_2Si_2$ is not due to a Cu deficiency to $\pm 5\%$. An order-parameter determination for our single crystals would be necessary to completely answer the question[7] of whether lattice perfection in $CeCu_2Si_2$ is essential for superconductivity.

ACKNOWLEDGMENTS

The authors wish to acknowledge valuable discussions with T. H. Geballe, M. B. Maple, and W. A. Harrison. We thank J. O'Rourke for performing the x-ray fluorescence measurement, L. Levinson for performing the energy-dispersive-spectroscopy measurement, D. Rohr for performing the electron-diffraction measurement to look for charge-density waves, and J. L. Smith and M. Wire for technical assistance. This work was performed under the auspices of the U. S. Department of Energy.

[1] F. Steglich, J. Aarts, C. D. Bredl, W. Lieke, D. Meschede, and W. Franz, Phys. Rev. Lett. 43, 1892 (1979).

[2] L. E. DeLong, J. G. Huber, K. N. Yang, and M. B. Maple, in *superconductivity in d- and f- Band Metals,* edited by W. Buckel and W. Weber (Kernforschungszentrum, Karlsruhe, 1982), p. 467.

[3] F. Steglich, J. Aarts, C. D. Bredl, G. Cordier, F. R. de Boer, W. Lieke, and U. Rauchschwalbe, in *Superconductivity in d- and f- Band Metals,* p. 145.

[4] W. Lieke, U. Rauchschwalbe, C. D. Bredl, F. Steglich, J. Aarts, and F. R. de Boer, J. Appl. Phys. 53, 2111 (1982).

[5] U. Rauchschwalbe, W. Lieke, C. D. Bredl, F. Steglich, J. Aarts, K. M. Martini, and A. C. Mota, Phys. Rev. Lett. 49, 1448 (1982).

[6] G. W. Hull, J. H. Wernick, T. H. Geballe, J. V. Waszczak, and J. E. Bernardini, Phys. Rev. B 24, 6715 (1981).

[7] C. D. Bredl, H. Spille, U. Rauchschwalbe, W. Lieke, F. Steglich, G. Cordier, W. Assmus, M. Herrmann, and J. Aarts, in International Conference on Magnetism, 1982, Kyoto, Japan [J. Magn. Magn. Mater. (in press)].

[8] S. Horn, E. Holland-Moritz, M. Loewenhaupt, F. Steglich, H. Scheuer, A. Benoit, and J. Flouquet, Phys. Rev. B 23, 3171 (1981).

[9] F. G. Aliev, N. B. Brandt, R. V. Lutsiv, V. V. Moshchalkov, and S. M. Chudinov, Zh. Eksp. Teor. Fiz. Pis'ma Red. 35, 435 (1982) [JETP Lett. 35, 539 (1982)].

[10] F. G. Aliev, N. B. Brandt, E. M. Levin, V. V. Moshchalkov, S. M. Chudinov, and R. I. Yasnitskii, Fiz. Tverd. Tela (Leningrad) 24, 289 (1982) [Sov. Phys.—Solid State 24, 164 (1982)].

[11] W. Reiger and E. Parthe, Monatsh. Chem. 100, 444 (1969).

[12] G. R. Stewart, Cryogenics 18, 120 (1978).

[13] G. R. Stewart and A. L. Giorgi, Phys. Rev. B 17, 3534 (1978).

[14] G. R. Stewart, B. Cort, and G. W. Webb, Phys. Rev. B 24, 3841 (1981).

[15] D. L. Martin (unpublished).

[16] T. H. Geballe (private communication).

[17] T. H. Geballe, B. T. Matthias, K. Andres, E. S. Fisher, T. F. Smith and W. H. Zachariasen, Science 152, 755 (1966).

[18] E. S. Fisher, T. H. Geballe, and S. M. Schreyer, J. Appl. Phys. 39, 4478 (1968).

[19] W. E. Gardner and T. F. Smith, Phys. Rev. 154, 309 (1967).

[20] For a review see S. D. Bader, N. E. Phillips, and E. S. Fisher, Phys. Rev. B 12, 4929 (1975).

[21] For a discussion, see G. H. Lander, in *Actinides in Perspective,* edited by N. M. Edelstein (Pergamon, New York, 1982) p. 107.

[22] J. C. Ho, N. E. Phillips, and T. F. Smith, Phys. Rev. Lett. 17, 694 (1966).

[23] T. G. Berlincourt, Phys. Rev. 114, 969 (1959); C. A. Cornelius and T. F. Smith, J. Low Temp. Phys. 40, 391 (1980).

[24]G. K. Bristow, T. R. Finlayson, and T. F. Smith, Phys. Status Solidi B 82, K81 (1977); G. K. Bristow, C. A. Cornelius, T. F. Smith, and T. R. Finlayson, J. Phys. F 8, 2165 (1978).

[25]J. R. Cullen, J. J. Rhyne, and F. Mancini, J. Appl. Phys. 41, 1178 (1970).

[26]W. Franz, A. Griessel, F. Steglich, and D. Wohlleben, Z. Phys. B 31, 7, (1978).

[27]For a discussion, see A. J. Heeger, in *Solid State Physics,* edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic Press, New York, 1969), Vol. 23, pp. 283—411. See particularly p. 358.

[28]There is some question as to the units used for this number. Judging from the Debye temperature quoted in Ref. 3 derived from β for $LaCu_2Si_2$, the intended value for γ may have been 4.4 mJ/g-at. K^2, or 22 mJ/mole K^2.

[29]Or perhaps unpublished data to higher temperatures were used.

[30]P. E. Bloomfield and D. R. Hamann, Phys. Rev. 164, 856 (1967).