Electrical and magnetic properties of the compounds $Ce_xLa_{1-x}Cu_2Si_2$ ($0 \leqslant x \leqslant 1$)

F. G. Aliev, N. B. Brandt, V. V. Moshchalkov, O. V. Petrenko, S. M. Chudinov, and R. I. Yasnitskiĭ

Moscow State University

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The electrical and magnetic properties of the substitutional solid solutions $Ce_xLa_{1-x}Ci_2Si_2$ ($0 \leqslant x \leqslant 1$) have been investigated in a broad temperature range ($0.04 \leqslant T \leqslant 300$ K) and magnetic fields $H \leqslant 40$ kOe. The T-X phase diagram has been constructed for the magnetic properties of the system $Ce_xLa_{1-x}Cu_2Si_2$. It is shown that these compounds become spin glasses at $T = T_{SG}$. The spin-glass transition temperature increases from about 330 mK ($x = 0.2$) to 1.6 K ($x = 0.6$). However, as the concentration of the magnetic component rises from $x \simeq 0.7$ to $x = 1$, the transition temperature T_{SG} falls, and the compound $CeCu_2Si_2$ becomes nonmagnetic and superconducting with $T_c \simeq 0.5$ K. This change of the magnetic properties of the solid solutions $Ce_xLa_{1-x}Cu_2Si_2$ is attributed to the transition from the state with $T_K < T_{RKKY}$ ($x \lesssim 0.7$), for which the magnetic transition characterized by the RKKY interaction temperature $T_{RKKY} \sim T_{SG}$ occurs before the Kondo compensation of magnetic moments (for $T < T_K$), to the state $T_K \gg T_{RKKY}$, for which the ground state of Ce^{3+} is a nonmagnetic singlet (T_K is the Kondo temperature). The Hall coefficient R_H of $CeCu_2Si_2$ is found to increase by a factor of 15–20 when the temperature is reduced from about 100 K to about 4 K. The ratio R_H (2.4 K)/R_H (70 K) and R_H (4.2 K) decrease with decreasing cerium concentration. The anomalous behavior of $R_H(T)$ shows that, as the temperature is reduced from $T \gg T_K$ to $T \ll T_K$, a narrow ($\sim kT_K$) resonance is formed at the Fermi level and corresponds to quasiparticles with very low degeneracy temperature. The onset of this resonance is shown to be responsible for the anomalous properties of Kondo lattices that are connected with the giant density of states at $\varepsilon = \varepsilon_F$. Studies of the superconducting properties of $CeCu_2Si_2$ at normal pressure and for pressures up to 13.4 kbar have shown that the giant values of the derivative of the upper critical field, $dH_{c2}/dT(T_c)$, and the fact that the paramagnetic limit is exceeded by a factor of two, are due to the formation of a resonance with an anomalously large amplitude at the Fermi level.

INTRODUCTION

In compounds containing rare-earth elements (REE), the localization radius of $4f$ states is very small ($r_{4f} \sim 0.2$ Å, see Ref. 1) and the width of the $4f$ band is only $\Delta \sim 0.1$–0.5 eV. Since the width of the s band is $W \sim 10$ eV, the probability that the $4f$ level will lie near the Fermi level ε_F is not too high ($\Delta / W \sim 0.5/10 = 0.05$), and compounds containing the REE usually have integer valence. For example, the valence of cerium in such compounds is [2] $\nu_{Ce} = 3$. However, there is a whole class of compounds[3] for which ε_F crosses the $4f$ level (these are the intermediate valence compounds). In intermediate valence compounds, the valence of the REE is not an integer and the mean time of ($4f^n \rightleftarrows 4f^{n-1} + s$) valence fluctuations is[3] 10^{-13} s. The fraction of $4f^n$ and $4f^{n-1}$ states is then of the same order of magnitude.

The states $Ce^4 + (4f^05d^26s^2)$, $Sm^{2+}(4f^65d^06s^2)$, $Eu^{3+}(4f^65d^16s^2)$, $Yb^{2+}(4f^{14}5d^06s^2)$ have zero total magnetic moment I, so that valence fluctuations are also spin fluctuations between magnetic and nonmagnetic states.

Since they are systems with charge fluctuations, the intermediate valence compounds occupy an intermediate position between purely localized states and free band states. Hence, intensive studies of systems with intermediate valence can, in principle, lead us to a unified description of the behavior of electrons in solids as the ratio of the degrees of localization to delocalization is varied.

In a certain sense, the intermediate-valence compounds are analogous to the states of impurity systems near the Mott-Anderson metal-dielectric transition, the analogy being confined to the immediate neighborhood of this transition. On the other hand, intermediate valence compounds are compounds with $I = 0 \rightleftarrows I \neq 0$ spin fluctuations and thus occupy a special position between the ordinary "stable" magnetism of dielectrics or metals and "nonmagnetic" solids. The magnetic properties of intermediate-valence compounds are therefore also of exceptional interest since they enable us to take the rare-earth ions from the magnetic to the nonmagnetic behavior through the "unstable" magnetism of the intermediate valence compounds.

This approach reflects the current tendency in solid-state physics toward a unified description of physical phenomena from one extreme, for which there is a solution, to another known limit, through all the "intermediate" values of the parameters that characterize the problem, using ideas based on the renormalization-group method, scaling theory, and so on.

Since, in compounds containing rare-earth elements, the localized magnetic moments associated with the $4f$ shell can interact through band states, the data obtained as a result of experimental and theoretical studies of the behavior of magnetic impurities in nonmagnetic metals are extensive-

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ly used in the physics of intermediate valence compounds. This problem (the Kondo problem) includes both the interaction of localized magnetic moments themselves through band states (RKKY oscillations characterized by energy $KT_{\rm RKKY} \sim J^2$, where J is the intensity of the antiferromagnetic interaction of $4f$ states with electrons in the conduction band) and the Kondo effect, which is due to the scattering of carriers by localized magnetic moments with spin flip and was the characteristic energy

$$kT_{\rm K} \sim \varepsilon_F \exp(-1/N(\varepsilon_F)J),$$

where $N(\varepsilon_F)$ is the density of states at ε_F. Complete dynamic compensation of the impurity magnetic moment by band electrons due to the formation at ε_F of a multiparticle Abrikosov-Suhl resonance[5–7] occurs below the temperature T_K.

Since $J \approx |V|^2/(\varepsilon_F - \varepsilon_{4f})$, where V is the matrix element of the s–f exchange interaction, the exchange interaction is small in the "usual" magnetic compounds based on rare-earth elements in which the $4f$ level lies well below the Fermi level, so that the temperature $T_{\rm K}$ that is an exponential function of $-1/N(\varepsilon_F)J$ will be negligible in comparison with the RKKY temperature, which is a quadratic function of J, i.e., $T_{\rm RKKY} \sim J^2$.

In this situation, the dynamic compensation of localized magnetic moments (Kondo effect) can have no appreciable effect on the magnetic transition, but should occur well before the onset of the compensation of local magnetic moments. Since the spins are locally frozen after the magnetic transition, the scattering of band electrons with spin flip is not possible.

The exchange integral J can be very large in intermediate-valence compounds because the $4f$ level is anomalously close to the filling limit. This means that the various external parameters, such as pressure, substitution of components resulting in "chemical compression," and so on, can be used in these materials to investigate the state of magnetic ions between $T_{\rm K} \ll T_{\rm RKKY}$ and $T_{\rm K} \gg T_{\rm RKKY}$. From this point of view, compounds in which the REE valence is still an integer, but the $4f$ level is so close to ε_F that the characteristic temperature $T_{\rm K}$ is high, occupy a separate class. These are the Kondo lattices, a phrase introduced by Doniach[8] for a one-dimensional chain representing the periodic distribution of a Kondo impurity.

However, the phrase "Kondo lattice," interpreted as "a periodic Kondo impurity," can be applied to any crystalline solid in which a $4f$ or $3d$ element is in a magnetic state. Nevertheless, for most solids in which the $4f$ level lies at a sufficient depth below ε_F, the new definition is inappropriate because $T_{\rm K} \ll T_{\rm RKKY}$ in such compounds and none of the effects connected with the dynamic compensation of local magnetic moments will appear, since a transition to a magnetically ordered state occurs at higher temperatures, and scattering with spin flip cannot occur on the locally-frozen spins. The use of the phrase, Kondo lattice, is therefore confined in the literature mainly to compounds of the form $CeAl_3$, $CeCu_2Si_2$, CeB_6, and $CeAl_2$, in which the $4f$ level lies immediately above the Fermi level, and which occupy an intermediate position between ordinary compounds containing a rare-earth element and the intermediate valence compounds. This usage of the phrase "Kondo lattice" will be adopted in this review.

The inequality $T_{\rm K} \gg T_{\rm RKKY}$ is satisfied for the $CeAl_3$ and $CeCu_2Si_2$ Kondo lattices, so that they are compounds that, for $T \to 0$, contain cerium in a nonmagnetic singlet state. On the other hand, $T_{\rm K} \lesssim T_{\rm RKKY}$ in both CeB_6 and $CeAl_2$, and these compounds exhibit a transition to a magnetic state on partially Kondo-compensated magnetic moments of the cerium ions. It is interesting that the compound $CeCu_2Si_2$ is not only nonmagnetic[9] as $T \to 0$, but also undergoes a transition to the superconducting state with anomalous parameters.[10–12] Among Kondo lattices and intermediate valence compounds, $CeCu_2Si_2$ is the only presently known superconductor and is therefore the subject of intense research at present.[9–21]

In the present paper, the transition from a magnetic state through "unstable" magnetism to a nonmagnetic state, and from integer valence of a rare-earth element through intermediate valence compounds to a state with unfilled $4f$ shell, is investigated by studying the example of isostructural substitutional solid solutions $Ce_xLa_{1-x}Cu_2Si_2$ $(0 \leqslant x \leqslant 1)$, in which an increase in the concentration of cerium is accompanied by a continuous transition from the Kondo impurity $(x \ll 1)$ to the Kondo lattice $(x \simeq 1)$, whereas the application of pressure results in a gradual transition of the $CeCu_2Si_2$ Kondo lattice with valence [21]$\nu_{\rm Ce} \simeq 3.03$ through the intermediate valence compound to the ordinary metal with $\nu_{\rm Ce} \simeq 4.0$ and unfilled $4f$ shell.[19]

The electrical and magnetic properties were investigated for pressures up to 140 kbar, magnetic fields up to 40 kOe, and low and ultralow temperatures (40 mK $\leqslant T \leqslant$ 300 K).

EXPERIMENTAL PROCEDURE. SPECIMENS

Infralow temperatures were produced with the aid of the He^3–He^4 refrigerator manufactured by the SHE Company in the United States. Magnetic fields were produced by a superconducting cable solenoid. Hydrostatic pressures up to 15 kbar were produced in a high-pressure chamber made of thermally treated BRB-2 bronze, using the Itskevich method. Pressures in the range $15 < p < 140$ kbar were produced in Bridgman anvils. For experiments at ultralow temperatures, the high-pressure chamber was joined to the refrigerator dilution chamber by means of a special cuprite holder. The temperature of the mixture in the dilution chamber was measured with a *GRT* germanium thermometer. Measurements in a magnetic field at intermediate temperatures were performed in a sealed cryostat.

The differential magnetic susceptibility was measured with a radiofrequency Zimmerman SQUID. The specimen was located in one of the coils of a superconducting transformer on the outer side of the dilution chamber. The technique is described in detail in Refs. 22 and 23.

Single-crystal $CeCu_2Si_2$ specimens and polycrystalline specimens (see the Table) were produced by direct fusing stoichiometric quantities of components in aluminum-oxide based ceramic crucibles, equipped with a tungsten heater. The crucible charge was maintained in the molten state for

TABLE I.

x	a, Å	c, Å	V, Å³	$T^{(1)}_{max\,\rho}$, K	A, $\mu\Omega\cdot$cm/K^2	T_{SG}, K	T_C, K
0	4,132	9,913	169,25	–	–	–	–
0,2	4,131	9,924	169,35	–	–	0,32	–
0,3	4,125	9,929	168,95	–	–		–
0,4	4,120	9,933	168,6	2.5	–		–
0.5	4,124	9.940	169,05	3,3	2	1,2	–
0,6	4,116	9.938	168.36	4.6	3,4	1,6	–
0.7	4.105	9,944	167.57	5,6	5,8	>2,5	–
0,8	4,101	9.958	167.48	6,9	5		–
0.9	4,100	9.950	167,26	7.9	5,4	2.2	–
1	4.099	9,924	166.7	8–10	9,3	–	0÷0,53
single crystal $CeCu_2Si_2$	4.101	9,924	166.9	6.4	7,4	–	<0.03 (p=0)

30 minutes and was then gradually cooled down. Alloys prepared in this way were subjected to homogenizing annealing at 800 °C for 150 hours. Composition and homogeneity were monitored by x-ray phase analysis and, for single crystal $CeCu_2Si_2$–by x-ray structural analysis.

VARIATION IN ELECTRICAL AND MAGNETIC PROPERTIES OF $Ce_xLa_{1-x}Cu_2Si_2$ IN THE COURSE OF A TRANSITION FROM A KONDO IMPURITY TO A KONDO LATTICE

$LaCu_2Si_2$ is a normal metal[10] so that the introduction of magnetic Ce^{3+} ions into this compound produces the Kondo rise in resistivity $\rho(T)$ (curves 1–6 in Fig. 1) with decreasing temperature:

$$\rho=\rho_p+\rho_0[1+\text{const}\cdot\ln(T/T_K)]x=\rho_p+\rho_m, \tag{1}$$

where ρ_b is the phonon contribution to resistivity, ρ_0 is the unitary limit for Kondo scattering, and ρ_m is the contribution due to the scattering of electrons by magnetic cerium ions. It is assumed in this expression that, for $T \gtrsim T_K$, the cerium atoms behave as independent scattering centers.

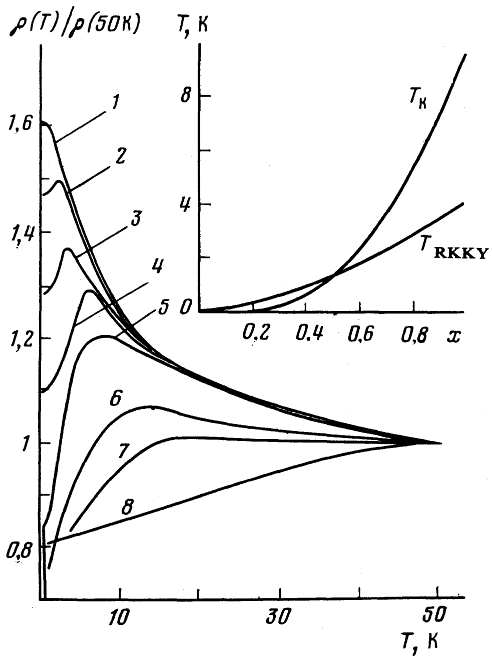

FIG. 1. Resistivity of the solid solutions $Ce_xLa_{1-x}Cu_2Si_2$ for different x (1—0.2; 2—0.4; 3—0.5; 4—0.7; 5—1) and $CeCu_2Si_2$ under pressure (6—10 kbar; 7—16.6 kbar; 8—45 kbar).

For concentrations $x \leqslant 0.3$, the saturation of ρ as $T \to 0$ is not accompanied by a reduction in resistivity ρ, at least for temperatures, $T \gtrsim 0.05$ K. Beginning with $x = 0.4$, the $\rho(T)$ curves exhibit a maximum (curves 2–6 in Fig. 1) at $T = T^0_{\max}$, where $T^0_{\max}$ increases with increasing cerium concentration along the sequence $Ce_xLa_{1-x}Cu_2Si_2$ at the rate $\partial T^0_{\max}/dx \approx 0.11$ K/at.% We note that the position of the maximum on the $\rho(T)$ curve of $CeCu_2Si_2$ is very sensitive to the degree of annealing. For polycrystalline specimens, $T^0_{\max}$ ($CeCu_2Si_2$) $\approx$ 8–10 K, whereas, for single crystals, this temperature is somewhat lower and amounts to 6.4 K.

An increase in x is also accompanied by a deviation of the reduced resistivity $\rho(T)/\rho(50\text{ K})$ from the common curve close to curve 1 of Fig. 1. This means that the temperature T_K of $Ce_xLa_{1-x}Cu_2Si_2$ alloys varies with increasing x. This variation can be approximately estimated as follows.

We shall suppose that T_K in (1) is a function $T_K(x)$ of the concentration. For example, if we compare curve 3 with curve 1 (Fig. 1), we can determine $T^{(1)}_K$ from the known $T^{(3)}_K$, using the relation $T^{(1)}_K = \overline{\alpha} T^{(3)}_K$. By averaging over different T, we can determine the coefficient $\overline{\alpha}$ for which $\rho(T)/\rho(50\text{ K})$ (curve 3 in Fig. 1) becomes superimposed on curve 1. This procedure for estimating $T_K(x)$ shows that the Kondo temperature increases as $x \to 1$ (insert in Fig. 1). The $T_K(x)$ curve was then normalized to $T_K(CeCu_2Si_2) = 10\ \kappa$, determined from the half-width of the neutron scattering line $\Gamma/2(T \to 0) \approx T_K \approx 10$ K (Ref. 9) and from measurements of $\chi(T)$ (Ref. 10).

The determination of $T_K(x)$ from the $\rho(T)$ curves (Fig. 1) is based on the assumption that scattering by localized magnetic moments in $Ce_xLa_{1-x}Cu_2Si_2$ is more effective than scattering by phonons. The phonon contribution ρ_p can be estimated from the shape of $\rho(T)$ for $LaCu_2Si_2$, where ρ(50 K) $\approx 25\ \mu\Omega\cdot$cm. Hence, it follows that, for $T < 50$ K, we have $\rho_m/\rho_p \geqslant 6$ and, consequently, the above procedure of estimating $T_K(x)$ is completely valid.

The low-temperature portion of the $\rho(T)$ curve corresponding to $T \to 0$ is described by $\rho = \rho_{\text{res}} + AT^2$. For $CeCu_2Si_2$ single crystals, $\rho_{\text{res}} \simeq 70\ \mu\Omega\cdot$cm and $A \simeq 7.4\ \mu\Omega\cdot$cm/K^2. Along the $Ce_xLa_{1-x}Cu_2Si_2$ series, the constant A is 9.3

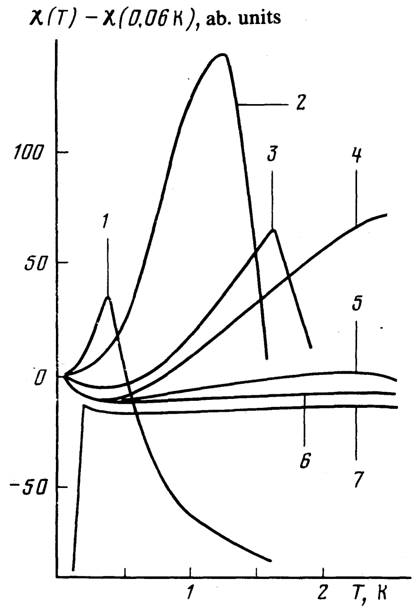

FIG. 2. Magnetic susceptibility $\chi(T) - \chi(0.06\ \mathrm{K})$ of $Ce_xLa_{1-x}Cu_2Si_2$ for different values of x (1—0.02; 2—0.5; 3—0.6; 4—0.7; 5—0.9; 6—$CeCu_2Si_2$ single crystal). Curve 7 corresponds to $\chi(T)$ for polycrystalline $CeCu_2Si_2$.

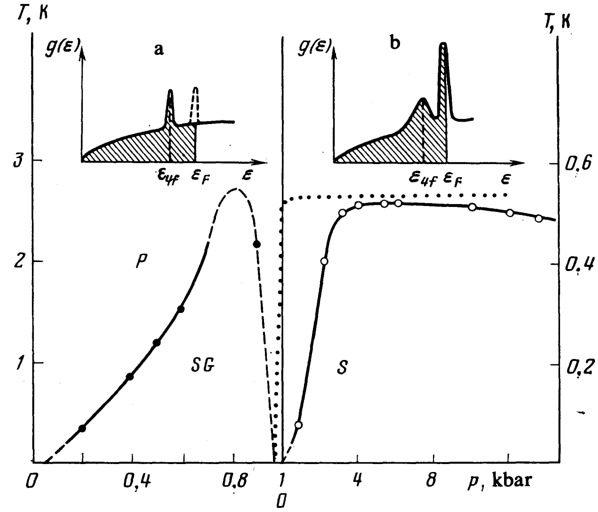

FIG. 3. Phase diagram for the magnetic and superconducting properties of the system $Ce_xLa_{1-x}Cu_2Si_2$ for $0 \leqslant x \leqslant 1$ and $0 < p \leqslant 13.4$ kbar (P—paramagnetic phase, SG—spin glass, S—superconducting phase). The function $T_c(x, p)$ is shown by the points and corresponds to Steglich's data.[14] Insets show the density of states $g(\varepsilon)$ for $T_K < T_{RKKY}$ (a) and $T_K > T_{RKKY}$ (b).

$\mu\Omega\cdot cm/K^2$ at $x = 1$ and falls to $2\mu\Omega\cdot cm/K^2$ for $x = 0.5$ (see the table).

The magnetic susceptibility χ (Fig. 2) of alloys with $0.2 \leqslant x \leqslant 0.6$ is a nonmonotonic function of temperature. The maximum of $\chi(T)$ corresponds to the transition to a spin glass at $T = T^{\chi}_{max} = T_{SG}$. The parametric behavior is observed for $T > T_{SG}$ and corresponds to the Curie-Weiss law $\chi = C/(T + \Theta)$. The temperature θ for the alloy with $x = 0.2$, for which the $\chi^{-1} = f(T)$ curve has the longest linear segment, is 180 mK.

As the concentration of the magnetic component increases, the paramagnetic (P)→spin glass (SG) transition temperature (Fig. 2) at first increases and then, beginning with $x \simeq 0.8$, T_{SG} exhibits an unusual behavior: an increase in the concentration of the magnetic component does not produce an enhancement of magnetism but, on the contrary, an effective suppression: the height of the maximum and T_{SG} are reduced (curve 5 in Fig. 2) and, for the maximum cerium concentration in the series $Ce_xLa_{1-x}Cu_2Si_2$, instead of the transition to the magnetically ordered state, one observes the superconducting transition (curve 7 in Fig. 2).

At the same time, as $x \to 1$, the paramagnetic behavior of $\chi(T)$ is substantially suppressed (see curves 5 and 6 in Fig. 2). The temperature-independent Pauli paramagnetism is observed for $x = 0.9$ and $x = 1$, and is replaced for $T < 300$ mK by a small rise in χ. The polycrystalline $CeCu_2Si_2$ specimen shows a strong diamagnetic behavior corresponding to the onset of superconductivity at $T = T_c \simeq 180$ mK.

The phase diagram showing the magnetic properties of the $Ce_xLa_{1-x}Cu_2Si_2$ solid solutions and constructed from measurements of $\chi(t)$ (Fig. 2) is given in Fig. 3. The P–SG phase boundary was determined from the position of the maxima of $T^{\chi}_{SG}(x)$. In the temperature range that we investigated ($0.04 \leqslant T \leqslant 2.5$), the alloy with $x = 0.7$ (curve 2 in Fig. 2) showed only a reduction due to the transition to the spin glass for $T > 2.5\ K$.

The upper limit of the temperature range was $T = 2.5\ K$ because of the use of the squid magnetometry that enabled us to measure $\chi(T)$ in very weak ($H \sim 0.1$ Oe) external magnetic fields.[23] The specimen under investigation was placed in a superconducting flux transformer whose connecting leads were screened by a tube that assumed the superconducting state for $T_c \lesssim 3$ K.

In contrast to single crystals,[11] polycrystalline $CeCu_2Si_2$ specimens[10] are superconductors even under normal pressure. This is related[11] to the presence in such specimens of intergrain stresses equivalent to the application of an external pressure $\Delta p \neq 0$. $CeCu_2Si_2$ single crystals become superconductors under pressure[11] (see also the right-hand side of Fig. 3, where S is the superconducting phase). According to Steglich[14] (dotted line in Fig. 3), the superconductivity of $CeCu_2Si_2$ polycrystalline specimens is suppressed by the addition of 2–3 at.% La. However, the transition from the spin glass to the superconductor near $x = 1$ in $Ce_xLa_{1-x}Cu_2Si_2$ was not investigated in detail in our experiments because we did not have polycrystalline $Ce_xLa_{1-x}Cu_2Si_2$ with low concentrations of lanthanum (the lowest nonzero concentration of lanthanum was $x = 10$ at.%).

We assumed that the RKKY interaction temperature was close to the spin glass transition temperature, and used the experimental P–SG boundary for $x < 0.7$ to determine $T_{RKKY}(x)$, which was then extrapolated to $0.7 \leqslant x \leqslant 1$ (see insert in Fig. 1).

The variation in the electrical (Fig. 1) and magnetic (Fig. 2) properties of the $Ce_xLa_{1-x}Cu_2Si_2$ alloys with increasing cerium concentration can be interpreted qualitatively as follows. Consider the ratio of the strength of the RKKY interaction between the Ce^{3+} ions with the strength of the Kondo interaction between Bloch band electrons and localized magnetic moments of cerium ions as x increases along the series $Ce_xLa_{1-x}Cu_2Si_2$.

Since $T^{ij}_{RKKY} \propto J^2/R^3_{ij}$, where R_{ij} is the separation between the ith and jth cerium ions in $Ce_xLa_{1-x}Cu_2Si_2$, the strength of the RKKY interaction will also increase with increasing x:

$$T^{ij}_{RKKY} \propto J^2(x)/R_{ij}^3 \propto xJ^2(x). \tag{2}$$

The fact that the measured $T_{SG}(x)$ (shown in Fig. 1) is superlinear for $x < 0.7$ shows that the increase in x is accompanied not only by an increase in T_{RKKY} due to the reduction in the mean separation R_{ij}, but also because of the increase in the parameter J. An analogous conclusion has already been reached above as a result of the analysis of the variation in the form of the temperature dependence $\rho(T)$ of $Ce_xLa_{1-x}Cu_2Si_2$. Since $J \sim |V_{sf}|^2/(\varepsilon_F - \varepsilon_{4f})$, the dependence of J on the concentration x may be due to a small shift of the $4f$ level toward the Fermi level and to a change in the overlap of the $4f$ state [see the inset showing $g(\varepsilon)$ in Fig. 3], which affects the value of J through the matrix element V_{sf}. The volume of the unit cell (see the table) of $CeCu_2Si_2$ ($V = 167.67$ Å^3) is slightly smaller than that of $LaCu_2Si_2$ ($V = 169.25$ Å^3).

The condition $T_K \gg T_{RKKY}$ is satisfied near $x = 1$ and a reduction in temperature is accompanied by the appearance of the singlet state of the localized magnetic moments of Ce^{3+}, which corresponds to the appearance of the Abrikosov-Suhl multiparticle resonance[5–7] at ε_F. As a result, transition from the magnetic ($\mu_{eff} = 2.62\mu_b$) to the nonmagnetic ($\mu_{eff} \to 0$) state of the Kondo impurities occurs well before the transition to the antiferromagnetic-type state or to the spin glass at $T \sim T_{RKKY}$ becomes possible. The magnetic transition is then completely suppressed as a consequence of the appearance of the resonance at ε_F [see the right-hand graph of $g(\varepsilon)$ in Fig. 3]. When $T_K \ll T_{RKKY}$ [left-hand $g(\varepsilon)$ insert in Fig. 3], the magnetic transition occurs earlier, and the Abrikosov-Suhl resonance does not appear at ε_F. Dynamic compensation ($\mu_{eff} \to 0$) does not then occur because the localized magnetic moments of cerium are locally frozen.

The increase in $T_K(x)$ along the series $Ce_xLa_{1-x}Cu_2Si_2$ can be connected both with the broadening of the $4f$ band with increasing Ce^{3+} concentration and with a certain shift of the midpoint of this band relative to the Fermi level [see $g(\varepsilon)$ inserts in Fig. 3]. Thus, the limiting compound $CeCu_2Si_2$ in the series $Ce_xLa_{1-x}Cu_2Si_2$ is a Kondo lattice with Ce^{3+} in the nonmagnetic singlet state as $T \to 0$, which is realized because $T_K \gg T_{RKKY}$. This inequality establishes the relationship between the two competing mechanisms, namely, the tendency toward a transition to the magnetically ordered state, and dynamic compensation of localized magnetic moments by band electrons and the associated tendency for the formation of the nonmagnetic singlet state as $T \to 0$.

MULTIFREQUENCY RESONANCE NEAR THE FERMI ENERGY IN THE KONDO LATTICE

The specific feature of the $CeCu_2Si_2$ Kondo lattice is that this compound constitutes the boundary between the normal metal, containing the periodically distributed magnetic impurities with $\mu_{eff} \neq 0$, and the nonmagnetic metal ($\mu_{eff} \cong 0$) that becomes a superconductor. In fact, fast spin fluctuations suppress localized magnetic moments in compounds such as $CeCu_2Si_2$ and $CeAl_3$. For $T \gg T_K$, the spin system can be discussed as in classical physics but, for $T \ll T_K$, the spin system behaves as a Fermi liquid.[24] On the phase diagram plotted on the T, W/J plane, the Kondo lattices lie in the critical region corresponding to the transition at $T = 0$ from Pauli paramagnetism to antiferromagnetism.[25] The specific features of this group of compounds are reflected in a number of anomalies in their thermodynamic and transport parameters. Measurements of the specific heat of $CeCu_2Si_2$ and $CeAl_3$ show that these materials have record values (for metals) of the electronic specific heat: ($CeCu_2Si_2$) ≈ 1000 mJ/mol·K^2 (Ref. 11), ($CeAl_3$) ≈ 1620 mJ/mol·K^2 (Ref. 26). The temperature dependence of magnetic susceptibility[10] indicates the supression of the localized magnetic moments of Ce^{3+} ions as the temperature is reduced from $T \gg T_K$ to $T \ll T_K$. These data are confirmed by neutron scattering experiments: $CeCu_2Si_2$ and $CeAl_3$ do not undergo a transition to a magnetically ordered state as $T \to 0$, at least for temperatures down to 20 mK (Refs. 9 and 27). The half-width $\Gamma/2$ of the quasielastic scattering line of $CeCu_2Si_2$ and $CeAl_3$ is a function of the square root of the temperature, which is characteristic for Kondo systems,[28] whereas $\Gamma/2$ ($CeCu_2Si_2$) $\simeq 1$ MeV (Ref. 9) as $T \to 0$. These data indicate that the spin fluctuation state undergoes a change from $T_{sf} \sim T_K \ll T$ at high temperatures, for which $\Gamma/2 \sim T^{1/2}$ and $\Gamma/2 \ll T$, to $T_{sf} \sim T_K \gg T$.

Below T_K, the resistivity is given by $\rho = \rho_{res} + AT^2$ (see the table), which is characteristic for a Fermi liquid.[26]

It is interesting that estimates of T_K for $CeCu_2Si_2$, performed by four independent methods, yield roughly the same result, namely,

$$T_K^\rho \approx T_K^\Gamma \approx T_K^\chi \approx T_K^\gamma \approx 10 \text{ K}.$$

where T^ρ_K was determined from the maximum on the $\rho(T)$ curve,[13,17] $T^\Gamma_K \simeq \Gamma/2(0)$ was found from neutron scattering data,[9] T^χ_K was deduced from the saturation temperature of magnetic susceptibility $\chi(T)$ and the transition to $\chi(T) = \text{const}$,[10] and measurements of specific heat for $T \to 0$ were used to obtain $T^\gamma_K = \pi^2 R/6\gamma$ (Ref. 29) where R is the gas constant.

We have carried out the first detailed measurments of the temperature dependence of the Hall coefficient $R_H(T)$ (Figs. 4, 5, and 7) and of the resistivity $\rho(T)$ (Fig. 1) for the system $Ce_xLa_{1-x}Cu_2Si_2$ under normal pressure and for the $CeCu_2Si_2$ Kondo lattice under pressures up to 140 kbar. A preliminary report of these results was published in Refs. 15 and 19. For $CeCuSi_2$, the Hall coefficient (Figs. 4 and 5) and $\rho(T)$ (Fig. 1) show a temperature dependence that is most unusual for metals. While in typical metals $R_H(T) = \text{const}$ at reasonable temperatures, and ρ decreases with decreasing T, the converse obtains in $CeCu_2Si_2$, namely, ρ varies slowly (by a factor of 1.5), whereas R_H increases rapidly (by a factor of 15–20) as the temperature is reduced from 100 K to 4 K. $R_H(T)$ becomes saturated for $T \leqslant T^*_F \approx 10$ K. This form of

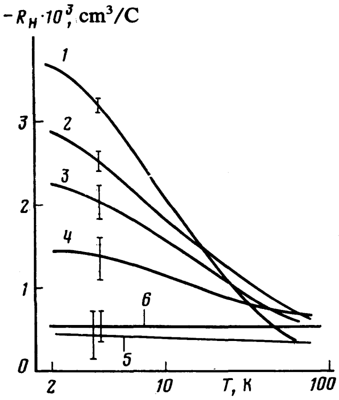

FIG. 4. Hall coefficient R_H of $Ce_xLa_{1-x}Cu_2Si_2$ for different values of x (1—1; 2—0.9; 3—0.5; 4—0.3; 5—0.2; 6—0).

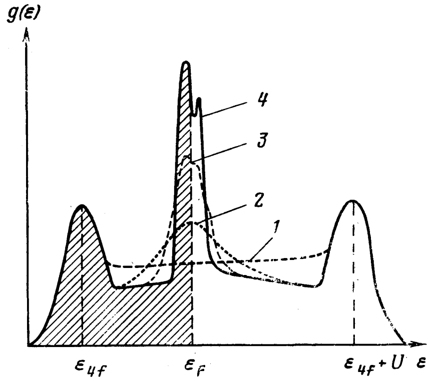

FIG. 6. Change in the energy spectrum of a Kondo lattice with varying temperature: $T \gg T_K$ (1), $T \sim T_K$ (2), $T \lesssim T_K$ (3), and $T \ll T_K$ (4).

the $R_H(T)$ curve indicates that there is a degeneracy temperature T_F^* above which nondegenerate Fermi statistics operates whereas, below this temperature, degenerate statistics must be employed. In normal metals, this degeneracy temperature amounts to $T_F \sim \varepsilon_F \approx 10\,000$ K, which lies above the melting point. A similar form of $R_H(T)$ is observed for doped semiconductors for which ε_F is anomalously low as compared with metals. Below T_F^*, which is also equal to T_K, the resistivity $\rho(T)$ and $R_H(T)$ show a behavior that is typical for normal metals for $T \to 0$: $\rho(T)$ falls and $R_H(T) = \text{const}$. Thus, $CeCu_2Si_2$ with free-electron concentrations $n(100\ K) \simeq 3 \times 10^{22}\ cm^{-3}$ behaves as a metal with extremely low degeneracy temperature for $T \lesssim T_K$, which is absolutely uncharacteristic for concentrations $10^{22}\ cm^{-3}$, for which $T_F \sim 1\ eV \sim 10\,000$ K.

What is the origin of this unusual behavior of $R_H(T)$ in $CeCu_2Si_2$ (Fig. 5)? When considered together with data on the specific heat,[10] magnetic susceptibility,[10] neutron scattering,[9] resistivity,[11,13] and the data listed in the table, the increase in $R_H(T)$ seems to us to indicate that the multiparticle Abrikosov-Suhl resonance[5–7] appears at ε_F when the temperature is reduced from $T \gg T_K$ to $T \ll T_K$ (Fig. 6). For a system of noninteracting localized magnetic moments, the appearance of this resonance at ε_F was first predicted by Abrikosov[6] and subsequently by Suhl,[7] who showed that the divergence exhibited by many perturbation theories as $T \to 0$ was connected precisely with the appearance of the multiparticle resonance at ε_F for $T \lesssim T_K$. The singularity in the density of states near ε_F was subsequently called the Abrikosov-Suhl resonance (see the review by Gruener and Zawadowski[30] on the Kondo problem). In particular, the Abrikosov-Suhl model has been used[30] to interpret the dependence of the residual resistance ΔR on the number N of $3d$ elements when they are dissolved in a given metallic host.

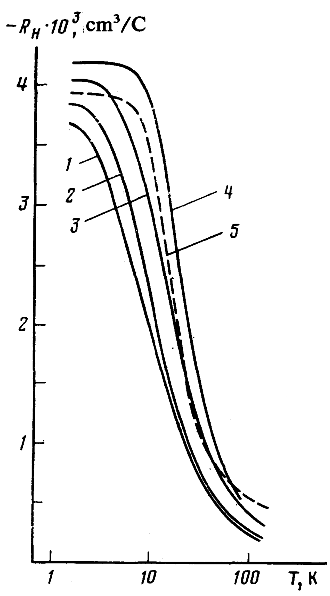

FIG. 5. Hall coefficient R_H of a $CeCu_2Si_2$ single crystal under different pressures (1—0.25 kbar; 2—0.9 kbar; 3—4 kbar; 5—9 kbar; 5—13.5 kbar).

The appearance of the multiparticle resonance was predicted theoretically in Refs. 31–33 for periodic Kondo systems. In particular, Martin and Allen[34] showed that, in contrast to an isolated impurity, periodicity in a system of Kondo impurities might lead to the appearance of a gap in the resonance at ε_F for a number of crystal directions in the Brillouin zone. The appearance of the gap is due to the requirement imposed on the volume of the Fermi surface, which should not change as the temperature is varied from $T \gg T_K$ to $T \ll T_K$, since the overall electron density in the system is constant.[34]

Despite the fact that the resonance at $\varepsilon = \varepsilon_F$ in concentrated Kondo systems is distorted by the presence of the correlation gap, the singularity in the interval $\sim kT_K$ near ε_F is found to remain and, in our view, is best referred to as the Abrikosov-Suhl resonance, just as in the case of an isolated Kondo impurity.

Since quasiparticles corresponding to excitations that form a Fermi gas for $T \to 0$ can be defined in a Fermi liquid[35] at sufficiently low temperatures, the effective mass of these quasiparticles can be estimated as follows. Knowing the density of states at the Fermi level[36] in terms of the coefficient γ, which is given by

$$N(\varepsilon_F) = 7.96 \cdot 10^{30}\ \gamma\,[\text{states/cm}^3\cdot\text{erg}\cdot\text{spin}], \quad (3)$$

where γ is in erg/cm³·K², and having determined the degeneracy temperature T_F^* from the $R_H(T)$ curve (Fig. 5), we can use the formula

$$N(\varepsilon_F)=4\pi(2m^*)^{3/2}\varepsilon_F^{1/2}/h^3 \qquad (4)$$

to show that $m^*\simeq 400m_0$ in the case of $CeCu_2Si_2$. Steglich *et al.*[12] previously reported a similar ($m^*\simeq 200m_0$) estimate for the effective mass of heavy fermions.

It is interesting that it is precisely the heavy fermions that are responsible for the onset of superconductivity in $CeCu_2Si_2$, since the jump in specific heat at $T=T_c$ corresponds to a jump in the anomalously large parameter γ that characterizes quasiparticles with large effective mass. The superconducting properties of $CeCu_2Si_2$ (Figs. 3, 8, and 9) are also anomalous: the ratio $H_{c2}(0)/T_c(0)$ for $CeCu_2Si_2$ is 38 kOe/K, which is double the paramagnetic limit.[37] The derivative of the upper critical field (Fig. 8) is 270 kOe/K, which is a record value for all known superconductors. The derivative $dH_{c2}/dT(T_c)$ can also be used to estimate the density of states $N(\varepsilon_F)$ since[36]

$$-dH_{c2}/dT\,(T=T_c)\,[\mathrm{Oe/K}]$$
$$=4.48\cdot 10^4\,\gamma\,[\mathrm{erg/cm^3K^2}]\cdot\rho\,[\Omega\cdot\mathrm{cm}]. \qquad (5)$$

The resulting value of $N_{\varepsilon F}$ is of the order of that determined from (3): $N_{\varepsilon F}(\gamma)/N_{\varepsilon F}(dH_{c2}/dT)\approx 2$.

The appearance of heavy fermions for $T\to 0$, in accordance with the calculations reported by Martin[31] (Fig. 1 of his paper), can be naturally related to the appearance of the multiparticle Abrikosov-Suhl resonance at ε_F. The energy spectrum near ε_F becomes greatly modified and a band[38] with an effective mass renormalized to a higher value can be introduced in the neighborhood of kT_K. Since the effective mass of heavy fermions in $CeCu_2Si_2$ is sufficiently high [$m^*\simeq(200\text{–}400)m_0$], the contribution of carriers with this effective mass will be negligible in the case of Hall-effect measurements (Figs. 4 and 5) in fields $H\leqslant 40$ kOe, and it may be considered that R_H is determined exclusively by the remaining light carriers with $m^*\lesssim m_0$. Since the resultant electron density $n=n(m^*\approx 200m_0)+n(m^*\lesssim m_0)$ is constant and temperature-independent, and $R\sim 1/n(m^*\lesssim m_0)$, the heavy-fermion concentration increases with increasing R_H. Thus, the increase in R_H for $T\to 0$ (Fig. 5) reflects the increase in the concentration of fermions with $m^*\gg m_0$ in $CeCu_2Si_2$ (Fig. 6).

It is interesting to note that the increase in R_H begins at the temperatures at which the logarithmic Kondo increase in the resistivity $\rho(T)$ appears (Fig. 1). We also note that the longest linear segments of $R_H=f(T)$ appear when the Hall coefficient $R_H(T)$ is plotted as a function of the logarithm of the temperature (Fig. 5) rather than the reciprocal temperature, as in the case of the activation-type dependence in doped semiconductors with $\varepsilon_F\sim 1\text{–}10$ meV. The straight-line dependence in the case of the logarithmic plot is in qualitative agreement with the data of Lacroix,[39] who calculated the density of states for the multiparticle resonance at $\varepsilon=\varepsilon_F$ as a function of temperature for the Kondo impurity (see Fig. 2 in Ref. 39):

$$n_R(\varepsilon_F)\approx n(\varepsilon_F)=f\left(1/\ln\frac{kT}{W}\right), \qquad (6)$$

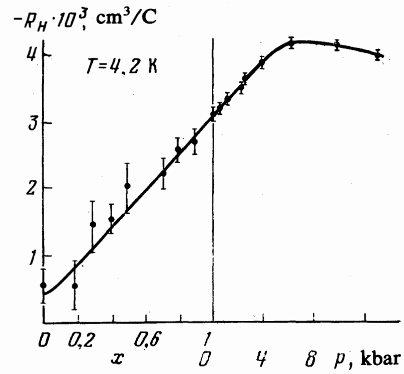

FIG. 7. Hall coefficient R_H (4.2 K) as a function of x for $Ce_xLa_{1-x}Cu_2Si_2$ and as a function of pressure p for the $CeCu_2Si_2$ single crystal.

where W is the bandwidth.

As the concentration of cerium in the $Ce_xLa_{1-x}Cu_2Si_2$ alloys (Fig. 4) is reduced, the graph of $R_H(T)$ is found to have a smaller slope, and R_H ($T\to 0$) decreases (Fig. 7). In $LaCu_2Si_2$ (curve 6 in Fig. 4), the Hall coefficient is independent of T. This behavior of R_H can be interpreted naturally as a reduction in the height of the multiparticle Abrikosov-Suhl resonance when the Ce^{3+} ions are diluted by the nonmagnetic La^{3+} ions.

In diluted Kondo systems and, in particular, in $Ce_xLa_{1-x}Cu_2Si_2$ with $x=1$, the height of the resonance at ε_F is small in comparison with the density of states $N^0(\varepsilon_F)$. On the other hand, an increase in the concentration x near $x\simeq 1$ is accompanied by the deformation of the function $N^0(\varepsilon_F)$, due to the appearance of the singularity near ε_F, so that (Fig. 6) the density of states $N^R(\varepsilon_F)$ that has been renormalized for $T\ll T_K$ is determined by the height of the resonance itself: $N^R(\varepsilon_F)\gg N^0(\varepsilon_F)$. For $x\simeq 1$, it is precisely the presence of the resonance at ε_F that is responsible for all the anomalous properties of Kondo lattices, namely, giant values of γ,

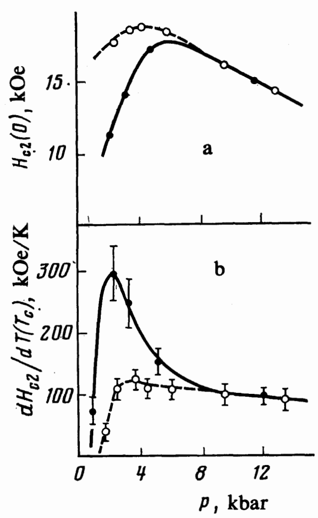

FIG. 8. Pressure dependence of the upper critical field H_{c2}(O) (a) and of the derivative dH_{c2}/dT ($T=T_c$) (b) for different directions of the magnetic field H and measuring current I: $\mathbf{H}\perp\mathbf{I}$(○) and $\mathbf{H}\parallel\mathbf{I}$(●).

anomalously high $dH_{c2}/dT(T=T_c)$, complete suppression of the magnetic moment ($\mu_{\text{eff}}\to 0$ for $T\to 0$), the increase in the Hall coefficient (Fig. 5), and the very low degeneracy temperature (Fig. 1,5).

Since the width of this resonance is [31] $\sim kT_K$ and its size in Kondo lattices is of the order of the concentration of magnetic ions, i.e., $\sim 10^{22}$ cm^{-3}, these two conditions can be satisfied for Kondo lattices only when the amplitude $N^R(\varepsilon_F)$ is large enough to ensure that γ and dH_{c2}/dT exceed the values characteristic for normal metals by a factor of two. At the same time, the small width and giant height of the resonance in Kondo lattices lead to low degeneracy temperature, which can be compared with the anomalously high effective mass ($m^*\simeq 200m_0$) of heavy fermions.

Thus, Kondo lattices essentially constitute a special class characterized by the presence of the tall and narrow Abrikosov-Suhl resonance at $\varepsilon=\varepsilon_F$ which, in turn, leads to a number of specific properties that can be seen experimentally, namely:

(1) suppression of the magnetic moments of rare-earth ions for $T\to 0$ ($T_K \gg T_{\text{RKKY}}$), and transition to enhanced Pauli paramagnetism;

(2) giant value of the electronic specific heat γ and of the derivative $dH_{c2}/dT(T_c)$ for a superconducting Kondo lattice (Fig. 8);

(3) anomalously low (for metals with $n\sim 10^{22}$ cm^{-3}) degeneracy temperature $T_F^*\sim T_K\sim 10$ K;

(4) high effective mass of heavy fermions that arises for $T\to 0$ in the transition from the interacting Fermi liquid to a Fermi gas of noninteracting quasiparticles corresponding to excitations;

(5) change in the sign of the thermal-expansion coefficient α (from $\alpha>0$ to $\alpha<0$) at $T\sim T_K$ (Ref. 24), so that the set of strongly interacting electrons in $CeAl_3$ and $CeCu_2Si_2$ with Grüneisen parameter $\Omega(T\to 0)=\Omega(CeAl_3)\approx -200$ can be looked upon[22] as a new example of a quantum-mechanical liquid analogous to He^3, where $\Omega(He^3)\approx -2$.

TRANSITION FROM THE KONDO LATTICE TO AN INTERMEDIATE VALENCE STATE IN $CeCu_2Si_2$ UNDER THE INFLUENCE OF PRESSURE

The separation $\overline{R}$ between the cerium ions is reduced as we proceed from $LaCu_2Si_2$ to $CeCu_2Si_2$ in which it reaches its minimum value for all the compounds in the series $Ce_xLa_{1-x}Cu_2Si_2$. However, if we use pressure as the external parameter, the magnetic ions in the $CeCu_2Si_2$ Kondo lattice can be brought even closer together.

When a pressure p is applied, the variation in the properties of the compound under investigation is initially the same as in the case of increasing x in $Ce_xLa_{1-x}Cu_2Si_2$ under normal pressure. For $0\leqslant p\leqslant 6$ kbar, the Hall coefficient $R_H(T\to 0)$ is found to increase (Fig. 7) with pressure. For a fixed value $p=p_0$, the coefficient R_H increases in the ratio of p to p_0 when the temperature is reduced from 100 K to 2 K (Fig. 5).

The graphs of $R_H^{T\to 0}(x)$ and $R_H^{T\to 0}(p)$ (Fig. 7) can be joined at the point $x=1$, $p=0$ in such a way that a single graph is obtained for R_H, first as a function of x and then as a function of p, without a break at $x=1$, $p=0$. It turns out that the ratio of the scales along the x and p axes is then the same as the ratio that can be obtained from the known shift of the maximum on the temperature dependence of resistivity $\rho(T)$ under the action of pressure[17] $dT^0_{\max}/dp\approx 0.7$ K/kbar, and as a result of a change in composition,[18] $dT^p_{\max}/dp\approx 0.11$ k/at.% (see the table), i.e., an increase in x by 0.1 is equivalent to a pressure of 1.6 kbar. By joining the curves at $x=1$, $p=0$, we obtain a ratio of the same order $\Delta x=0.1\leftrightarrow\Delta p=1.2$ kbar and, consequently, the variation in T^p is correlated with the function $R_H(x,p)$.

At the same time, for $0<p<3$ kbar, there is a sharp $dT_c/dp\approx 0.25$ K/kbar) increase in the transition temperature T_c to the superconducting state (see Fig. 3).

Since a hydrostatic pressure will shift the $4f$ level in the upward direction relative to ε_F, the Kondo temperature that is governed by the difference $\varepsilon_F-\varepsilon_{4f}$ will increase for $CeCu_2Si_2$ when the pressure is applied, just as when x is increased (see Fig. 1). The increase in R_H (4.2 K) (Fig. 7) can then be interpreted as a further increase in the weight $N^R(\varepsilon_F)$ of the Abrikosov-Suhl resonance and a reduction in the fraction of free electrons that do not "stick to" the Ce^{3+} ions. It is then natural to try and associate the abrupt rise in T_c under pressure with the increase in T_K.

During the dynamic compensation of localized magnetic moments in the Kondo effect, the reduced magnetic moments of the Ce^{3+} ions will prevent the formation of Cooper pairs[40] and the superconducting transition will occur only when the residual magnetism due to Ce^{3+} and the electrons "sticking" to it is negligible: $T_K\gg T_c$. The increase in T_c may then be visualized as a process in which the superconducting transition temperature follows the increase in the Kondo temperature: $dT_K/dp\sim dT_c/dp$. Estimates show that

$$dT_K/dp=T_K^{p=0}\frac{J^{p=0}}{\rho J}\frac{dJ}{dp},\quad dJ/dp=-\frac{V^2}{\Delta^2}\frac{d\Delta}{dp}. \tag{7}$$

If we take $\Delta\simeq 0.5$ eV (Ref. 2), $\varepsilon_F-\varepsilon_{4f}\sim 1$ eV (Ref. 2), and assume that $\varepsilon_{4f}=\varepsilon_F$ at 50 kbar, we find that $dT_K/dp\sim 1$ K/kbar. The experimental result is $dT_c/dp\simeq 0.25$ K/kbar, which is of the same order as the theoretical estimate.

This parallelism between the increase in T_K and the variation in the superconducting transition temperature will continue only up to the value T_c^0 that is observed for $CeCu_2Si_2$ with completely suppressed Ce^{3+} magnetic moments. The measured function $T_c(p)$ (Fig. 3) shows that, in $CeCu_2Si_2$, $T_c^0\sim 0.5$ K.

Further increase in all-sided pressure will induce a transition in $CeCu_2Si_2$ from the Kondo-lattice regime to an intermediate valence compound, for which the valence of the $4f$ state is not an integer, and the theoretical description of the behavior of the electron system must involve the Anderson Hamiltonian[34] and not the Kondo Hamiltonian.[8] By analogy with the Kondo lattice, the intermediate valence state itself can be referred to as the Anderson lattice.[34] *The Kondo* increase in $\rho(T)$ (Fig. 1) is then replaced by a metal-type reduction in resistivity with decreasing temperature for $T<50$ K and $p>40$ kbar. The transition from the Kondo lattice to the intermediate valence compound can also be induced by varying the composition, for example, by increasing[41] the component x in $CeIn_{3-x}Sn_x$.

In accordance with the theoretical calculations of Lacroix,[39] the transition from the Kondo impurity to the intermediate valence compound has associated with it a critical value of the difference

$$|\varepsilon_F-\varepsilon_{4f}|_c=\frac{\Delta}{\pi}\left(1-\ln\frac{\Delta}{\pi W}\right)=\Delta\varepsilon_c, \qquad (8)$$

which corresponds to the complete suppression of the Abrikosov-Suhl resonance at $\varepsilon=\varepsilon_F$. For $|\varepsilon_F-\varepsilon_{4f}|<\Delta\varepsilon_c$, the singularity at ε_F is less well defined as compared with the Kondo lattice, and decreases as $\varepsilon_{4f}\to\varepsilon_F$ to $N^R(\varepsilon_F)=0$ when $|\varepsilon_F-\varepsilon_{4f}|=\Delta\varepsilon_c$.

According to (8), $\Delta\varepsilon_c\simeq 0.7$ eV which, for $d|\varepsilon_F-\varepsilon_{4f}|/dp\approx 20$ meV/kbar, gives $p_c\simeq 20$ kbar if we use $p(\varepsilon_F=\varepsilon_{4f})\simeq 50$ kbar (Ref. 19) and $(\varepsilon_F-\varepsilon_{4f})\sim 1$ eV (Ref. 2). According to this estimate, the Abrikosov-Suhl resonance should vanish for $p\gtrsim p_c$. Regions of decrease in R_H (4.2 K) (Fig. 7) and in $T_c(p)$ (Fig. 3) can be naturally related to a reduction in the height of the resonance at ε_F since $CeCu_2Si_2$ superconductivity is, in fact, due to the superconductivity of heavy fermions that correspond to the appearance of the singularity in the density of states at ε_F.

The resonance may acquire a structure in the case of periodic Kondo lattices because of the appearance of a correlation gap[34] in a number of directions in the Brillouin zone. The anisotropy of the superconducting characteristics (Fig. 8) observed for $p\leqslant 6$ kbar may be connected with this particular fact. The anisotropy of dH_{c2}/dT for H_{c2} for H‖I H⊥I vanishes for $p\gtrsim 6$ kbar. This corresponds roughly to the beginning of the transition from the Kondo lattice to the intermediate valence compound, and the reduction in $dH_{c2}/dT(T_c)$ (Fig. 8) and, hence, in the density of states $N(\varepsilon_F)$ at the Fermi level, may be due to a reduction in the correlation gap under pressure.

Since the mean separation between the cerium ions in the layer perpendicular to the crystallographic c axis (see the table) is much greater than the separation between the layers, the large change in the derivative $dH_{c2\parallel}/dT$ under pressure (Fig. 8) as compared with $dH_{c2\perp}/dT$ indicates the unit cell has been subjected to a larger compression along the c axis than in the perpendicular direction.

Graphs of the critical fields $H_{c2\parallel}(T_c)$ (Fig. 9a) have a large slope at T_c ($H=0$). Graphs plotted on the logarithmic scale (Fig. 9b) show that

$$H_{c2}=H_{c2}(0)\left[1-\left(\frac{T_c(H)}{T_c(0)}\right)^{\alpha}\right], \qquad (9)$$

where $\alpha\simeq 4$–8, whereas, usually, $\alpha=2$.

Final conclusions with regard to the nature of superconductivity in $CeCu_2Si_2$ cannot as yet be deduced from existing experimental data (Fig. 3,8). However, from our point of view, these data indicate that: (1) the increase in T_c (Fig. 3, $p\leqslant 4$ kbar) is connected with the increase in T_K under pressure; (2) an increase in pressure for $p\leqslant 6$ kbar leads to an increase in the height of the multiparticle resonance at $\varepsilon=\varepsilon_F$ (heavy fermion concentration) and is responsible for

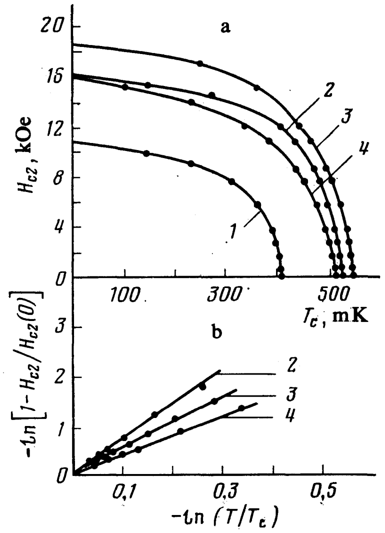

FIG. 9. Critical field $H_{c2\parallel}(T_c)$ (a) and the function $\ln[1-H_{c2}/H_{c2}(0)]=-\alpha\ln(T/T_c)$ (b) for different pressures: 1—2.2 kbar; 2—3.2 kbar ($\alpha\simeq 8$); 3—5 kbar ($\alpha\simeq 6$); 4—12 kbar ($\alpha\simeq 4$).

the increase in the Hall coefficient R_H (4.2 K) under pressure; and (3) the reduction in the anisotropy of dH_{c2}/dT and H_{c2} (Fig. 8) under pressure is due to the high compressibility along the c axis and the associated change in the correlation gap.

Thus, the transition from stable to unstable magnetism, which is, at the same time, a transition from localized $4f$ states to delocalized states, is found to occur in the system $Ce_xLa_{1-x}Cu_2Si_2$ when the position of the $4f$ level relative to ε_F is gradually altered by varying the composition and pressure. However, from our point of view, the most interesting result of our work is the discovery of the presence, just before the appearance of the intermediate valence compounds ($\varepsilon_F\simeq\varepsilon_{4f}$), of a new state of the Kondo-lattice type. This state is connected with a radical rearrangement of the density of states in the neighborhood of ε_F as the temperature is reduced from $T\gg T_K$ to $T\ll T_K$, and to the formation of the multiparticle Abrikosov-Suhl resonance at ε_F.

The height of this resonance is anomalously large, whereas its width is determined by T_K and amounts to 10 K in $CeCu_2Si_2$. It is precisely this singularity in the density of states that is responsible for the anomalous properties of the Kondo lattices, namely, giant values of the electronic specific sheet, suppression of magnetism for $T\to 0$, the onset of superconductivity with a number of anomalous propertie s, and the exceedingly low degeneracy temperature which, together with the high electron concentration, indicates that the heavy-fermion quasiparticles have an anomalously high mass.[12] The results reported here confirm the suggestion made by Steglich,[12,14] that the superconducting properties of $CeCu_2Si_2$ are due to the bonding into Cooper pairs of precisely the quasiparticles that correspond to the resonance at $\varepsilon=\varepsilon_F$.

In conclusion, we would like to take this opportunity to acknowledge our indebtedness to A. A. Abrikosov, D. I. Khomskiĭ, A. I. Buzdin, and R. V. Lutsiv for discussions of

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