

Nonmagnetic Kondo lattices

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Usp. Fiz. Nauk **149**, 585–634 (August 1986)

We present a discussion of the present state of experimental and theoretical investigations of a new class of metallic systems based on f -shell elements: the class of nonmagnetic Kondo lattices, in which the low-temperature density of electron states at the Fermi level is 100–1000 times larger than in normal metals, as a consequence of the generation of an astonishingly narrow ($\sim 10^3$ K) Abrikosov-Suhl resonance of gigantic amplitude near the Fermi level. This resonance is related to the presence of quasiparticles with effective masses $m^* \sim (10^2-10^3)m_0$ —“heavy fermions.” We show that the overwhelming majority of “heavy fermion systems” are in fact nonmagnetic Kondo lattices. We analyze the unusual properties of heavy-fermion superconductors, and point out the nontrivial nature of superconductivity in these systems.

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1. INTRODUCTION

Recently, much interest has attached to investigations of metallic systems with “heavy fermions.”¹⁻³ Typical representatives of such systems are various intermetallic compounds based on cerium ($CeAl_3$, $CeCu_2 Si_2$, $CeCu_6$) and uranium (UBe_{13} , UPt_3), which are endowed with a whole range of unusual low-temperature properties. The available experimental data from investigations of HFS suggest that at low temperatures the electronic density of states at the Fermi level $g(E_F)$ is amplified by roughly a factor of 10^2-10^3 compared to normal metals, i.e., the function $g(E)$ has a narrow peak ($\sim 10^3$ K) of gigantic amplitude in the vicinity

of the Fermi level. Since the characteristic band width in metals amounts to 1–10 eV (10^4-10^5 K), while the electron effective mass m^* lies within the limits $(0.1-1)m_0$, this astonishingly narrow band in HFS corresponds to “heavy fermions”—quasiparticles with effective masses $m^* \sim W^{-1} \sim (10^2-10^3)m_0$, where m_0 is the free electron mass.

What is the nature of this unusual feature in the function $g(E)$ for $E \approx E_F$? Where do heavy fermions come from in metals? These questions have recently occupied the lion's share of attention among those researchers who are engaged in the study of HFSs. For this reason, we have chosen as the fundamental goal of this review the elucidation of recently-acquired data which is directly relevant to clarifying the na-

ture of this gigantic peak in the HFS density of states which appears near the Fermi level.

All well-known HFSs are synthesized on the basis of $4f$ or $5f$ elements, which contain well-localized f -shells with uncompensated local magnetic moments. The characteristic radius r_f of these shells is extremely small, $0.3\text{--}0.5 \text{ \AA}$.⁴ Therefore, the direct overlap of f -shells is negligibly small even in the "concentrated" case when $4f$ or $5f$ elements are found in every unit cell. A density which corresponds to close-packing of $4f$ ($5f$) centers, i.e., at which the f -shells overlap, is unachievable under normal conditions, since this would require a concentration of $N_f \sim r_f^{-3} \sim 10^{25} \text{ cm}^{-3}$, which exceeds Avogadro's number N_A normalized to unit volume.

Localization of the magnetic moments of the $4f$ ($5f$) ions is quite apparent from various magnetic measurements made at low temperatures. It would seem that as the temperature falls, one ought to expect a transition to a magnetically ordered state in HFSs. However, in actuality a decrease in temperature leads to compensation of the effective magnetic moment $\mu_{\text{eff}}^2 \sim \chi T$ and to a nonmagnetic ground state in CeCu_2Si_2 ,⁵ CeAl_6 ,⁶ and other HFSs. Furthermore, in the compounds CeCu_2Si_2 ⁵ and UBe_{13} ,⁷ in place of a magnetic transition one observes a transition to a *superconducting state* at low temperatures; the same heavy fermions which give rise to the narrow peak in the density of states at E_F are also responsible for the superconductivity. An isomorphic nonmagnetic homologue of CeCu_2Si_2 , the compound LaCu_2Si_2 , which contains no magnetic ions (since the $4f$ shell of La^{3+} is empty) does not undergo a transition to the superconducting state.^{5,8} Comparing the isomorphic compounds CeCu_2Si_2 and LaCu_2Si_2 , we are thus led to an at first glance paradoxical conclusion: the presence of the magnetic ions Ce^{3+} in CeCu_2Si_2 favors the appearance of superconductivity. Whereas in magnetic superconductors,⁹ superconductivity is present under appropriate conditions in spite of the presence of magnetic ions, superconductivity in HFSs is present *because of* the presence of $4f$ ($5f$) ions which at high temperatures have uncompensated magnetic moments. This example illustrates how important the discovery of superconductivity in HFSs^{5,7,10} is to the study of the interplay between superconductivity and magnetism.

Superconductivity in the HFSs CeCu_2Si_2 , UBe_{13} and UPt_3 ,^{5,7,10} which is present under conditions of giant enhancement of the density of states at the Fermi level, is in a peculiar way analogous to superfluidity in ^3He .¹¹⁻¹⁴ Therefore, an interesting possibility suggests itself—that of using results obtained for ^3He (taking into account the limitations imposed by symmetry of HFSs) to describe the properties of a coherent magnetic metallic system in which heavy-fermion superconductivity is present. Thus, it would seem that the superconductivity of Kondo lattices provides an example of the merging of two totally disparate areas of low-temperature physics: the physics of quantum liquids and the physics of metals. Comparatively recently, a number of authors¹¹⁻¹⁵ have formulated the hypothesis that triplet pairing can occur in superconducting HFSs, in particular in the compound UPt_3 .¹⁵

These unusual properties of HFSs have aroused great interest among researchers around the world, and already today the volume of information on HFSs is growing rapidly, which makes it possible for us to separate out a few fundamental approaches to the study of HFSs.

One of the directions taken in studying the physics of HFSs relates to the investigation of compounds of the types CeCu_2Si_2 , CeAl_3 and UBe_{13} , which are treated as concentrated Kondo systems,^{2,16,17} in which the unusual feature in the density of states $g(E_F)$ is related to Kondo spin fluctuations mediated by electronic states which are close to the Fermi level and which lead to a sharp "weight gain" of the Fermi electrons. In the framework of this approach, the peak in $g(E_F)$ is in fact a giant Abrikosov-Suhl resonance (ASR).¹⁸⁻²⁰ The periodic CKS is also called a Kondo lattice. In the present review, we will focus our attention on those HFSs which can be treated as nonmagnetic Kondo lattices (NKLs).

Another group of papers has focused attention on investigating the Fermi-liquid properties of heavy fermions, in particular the possibilities for realizing unusual kinds of superconductivity without being interested (at least initially) in the process whereby the peak in $g(E)$ appears for $E \approx E_F$.

Finally, the unusual combination of low-temperature HFS properties may with some (apparently very small) probability be related to the superposition of a number of factors, each of which taken separately is quite ordinary and does not excite much interest. Such an approach, which contributes to the physics of HFSs a certain amount of skepticism and doubt,²¹ is a necessary element to the progress of development not only of the physics of HFSs but also of any other branch of science.

Let us now turn to an exposition of the arguments in favor of discussing the compounds CeCu_2Si_2 , CeAl_3 and UBe_{13} as NKLs. First of all, these compounds have extensive regions of logarithmic growth in the resistivity $\rho(T)$ at low temperatures, low-temperature suppression of the effective magnetic moment ($\mu_{\text{eff}}^2 \sim \chi T \rightarrow 0$ for $T \rightarrow 0$), and anomalous thermoelectric powers and heat capacities, which are all characteristic of the behavior of magnetic impurities in normal metals.²²⁻²⁴

The history of study of the behavior of isolated magnetic impurities dissolved in normal metals had its beginnings in the 1930s when in the course of investigating the low-temperature resistivity of metals researchers observed a minimum in $\rho(T)$ for $T = T_{\text{min}}$.²² Later it was established that the appearance of this minimum in a region where one expected a normal "metallic" decrease in $\rho(T)$ was related to the presence in these metals of magnetic impurities, and that the increase in the resistivity in the region $T < T_{\text{min}}$ could be described by a logarithmic function $\Delta\rho \sim -\ln T$.^{24,25} In 1964, Kondo²⁶ showed for the first time that logarithmic growth in the resistivity with decreasing temperature was a consequence of spin-flip scattering of sd electrons by the local magnetic moments, and could be obtained within perturbation theory by studying the second Born approximation. However, for the temperature

$$T \rightarrow T_K \sim T_F \exp \left[-\frac{1}{g(E_F) J \nu} \right] \quad (1.1)$$

(T_F is the Fermi temperature, T_K is the Kondo temperature, J is the exchange interaction constant and ν is the degeneracy of the magnetic level) the perturbation theory used by Kondo diverges. This divergence is a consequence of the creation at low temperatures of a narrow ($\sim k_B T_K$) many-particle Abrikosov-Suhl resonance near E_F ,^{18-20,27-29} corresponding to a transition from weak interactions between the band electrons and magnetic impurities at high temperatures $T \gg T_K$ to strong interactions at low temperatures $T \lesssim T_K$; in this temperature range, a quasibound state forms around the magnetic impurity—a “halo” of band electrons of oppositely-directed spins. In the region $T \ll T_K$, the Kondo process can be described using a picture based on Fermi liquid theory.³⁰⁻³¹

In the past year, an exact solution to the Kondo problem has been found by Vigman³² and Andrej,³³ based on the Bethe ansatz, which makes it possible to “fit together” all previous solutions by using a single function found by these authors.^{18-20,26-31} The Vigman-Andrej result is applicable to all temperature intervals, from $T \gg T_K$ to $T \ll T_K$. For low temperatures, i.e., $T \lesssim 10 T_K$, the dependence of the magnetic susceptibility $\chi(T)$ and heat capacity $C(T)$ as determined by the Vigman-Andrej solution can be approximated with good accuracy by a calculation based on the assumption that a Lorentzian resonance is present in $g(E)$ of width $\sim T_K$, which appears at the Fermi level.^{34,35} Thus, the transition (as a result of Kondo spin fluctuations) from magnetic (for $T \gg T_K$) to the singlet nonmagnetic impurity state as $T \rightarrow 0$ must be connected with the appearance of the narrow many-particle ASR near E_F . The amplitude of this resonance for one impurity equals $g_1^R(E_F) = 1/\pi\Delta_f$ (Δ_f is the hybridization width of the f level, which is located at $E = E_f$). As the temperature decreases in the range $T > (10-100) T_K$, the ASR is washed out²⁾ and a transition takes place to the original unperturbed density of states $g^0(E)$.³⁴⁻³⁷

Whereas the ASR amplitude $g_1^R(E_F)$ of a single magnetic impurity is negligibly small compared to the noninteracting density of states $g^0(E_F)$ in the sd band, when the concentration N_i of noninteracting magnetic impurities is large enough (comparable to that of the nonmagnetic atoms), the opposite situation obtains:

$$g_{N_i}^R(E_F) \sim \frac{N_i}{k_B T_K} \gg \frac{N_A}{W} \sim g^0(E_F). \quad (1.2)$$

The low-temperature density of states in a CKS with $N_i \sim (0.1-1.0) N_A$ and $T_K \sim 10$ K must therefore increase by a factor of $W/(k_B T_K) \sim 10^2-10^3$. However, to realize the ratio (1.2) in practice entails some definite problems. First of all, to create a CKS based on $3d$ elements is apparently impossible, since the radius of the $3d$ shell is rather large, and before any noticeable enhancement of $g_{N_i}^R(E_F)$ arises the interaction of the $3d$ shells is “switched on”; more precisely, a quasibound electron state forms around these shells. This problem can be avoided if we pick magnetic impurities consisting of $4f$ ($5f$) elements, in view of the extremely small radii of the localized f -shells.

Secondly, it is necessary to overwhelm in some way the indirect spin-spin interaction mediated by Ruderman-Kittel-Kasuya-Yosida (RKKY) oscillations in the spin density,³⁸⁻⁴⁰ because the Kondo spin fluctuations which lead to the formation of the ASR cannot occur in a system of spins which are “frozen” as a result of a magnetic transition. The intensity of the indirect RKKY interaction is characterized by a temperature T_{RKKY} :

$$T_{\text{RKKY}} \sim \frac{J^2}{W}. \quad (1.3)$$

The constant J in equation (1.3),³⁾ in its turn, depends on the degree of hybridization V_{sf} of the sd and f states, and the position of the f level relative to the Fermi level:

$$J \sim \frac{V_{sf}^2}{E_F - E_f}. \quad (1.4)$$

The ratio (1.4) was obtained by Schrieffer and Wolff⁴¹ for the asymmetric Anderson model⁴² with $(V_{sf}, E_F - E_f) \ll U$, where U is the repulsive Coulomb energy of two electrons with opposite spins in the magnetic field.

In the majority of intermetallic compounds based on $4f$ ($5f$) elements, the f level is located quite far below E_F and the constant J is extremely small. For small J , the Kondo temperature T_K (1.1), which depends exponentially on $(-1/J)$, is negligibly small compared to the temperature T_{RKKY} , which depends quadratically on J (1.3). In this case, as the temperature decreases, effects related to the formation of an ASR cannot come about, because a magnetic transition occurring at $T \sim T_{\text{RKKY}} \gg T_K$ “freezes” the magnetic moments and “forbids” the Kondo spin-flip processes. Thus, in order to obtain a CKS, it is necessary for the f level to be located directly beneath the Fermi level. In this case, the smallness of the denominator in (1.4) and a sufficiently large value of the numerator will guarantee a large value of the parameter J . In this situation the condition $T_K \gg T_{\text{RKKY}}$ is fulfilled and the magnetic transition turns out to be suppressed by the effectiveness of the Kondo spin-compensation process, and for sufficiently low temperatures an ASR forms with an amplitude enhanced N_i times compared to the resonance created by an isolated impurity. The arguments we will present show that the NKL is not simply a periodic system of Kondo centers, but rather is a periodic system of Kondo centers for which the conditions $T_K \gg T_{\text{RKKY}}$ holds, i.e., in a NKL the number of magnetic ions per mole must be so large that condition (1.2) is satisfied; however, in this case the indirect magnetic RKKY interaction between magnetic ions must also be suppressed.

For the NKLs CeAl_3 , CeCu_2Si_2 and UBe_{13} the approximation of an ensemble of noninteracting Kondo impurities turns out to be fairly good.¹⁶ This allows us to take advantage of the well-known results of the exact Vigman-Andrej solution³²⁻³³ in calculating the molar susceptibility $\chi(0)$ and coefficient γ of the electronic heat capacity (as $T \rightarrow 0$); one then simply multiplies χ and γ by Avogadro's number $N_A = N_i$. Historically, however, it turned out that the study of the NKLs CeAl_3 , CeCu_2Si_2 , etc., was approached not by treating the phenomenon as a natural consequence of an increase in the number of Kondo impurities but rather by re-

garding these compounds as variants of intermediate-valence compounds (IVCs).^{43,44} In these compounds the $4f$ band falls exactly at the Fermi level, and because of the interconfigurational transition $4f^n \rightleftharpoons 4f^{n-1} + sd$ (here n is the number of electrons in the $4f$ shell) a non-integral filling of the f -shell takes place. For IVCs, it is assumed that the narrow peak in the density of states near E_F is the $4f$ band itself. This interpretation was at first advanced also for the NKLs CeAl_3 and CeCu_2Si_2 . However, it was later established that the valence of cerium in CeAl_3 and CeCu_2Si_2 is close to integral $\nu(\text{Ce}) \approx 3.03-3.05$,⁴⁵⁻⁴⁷ and the unusual feature in the density of states in these compounds at the Fermi level could in no way be related directly to the f band itself. In Refs. 48 and 49 the suggestion was first explicitly made that in CeAl_3 and CeCu_2Si_2 the narrow peak in the density of states at the Fermi level is in fact a many-particle ASR, while the f band itself lies somewhat farther below E_F in NKLs and this corresponds to the integral valence $\nu(\text{Ce})$.

The width of the ASR is determined by the Kondo temperature T_K , while its position E_R relative to E_F depends on the total magnetic moment j of f electrons. The occupation of the ASR at $T \sim 0$ equals $1/(2j+1)$,³¹ i.e., for $j = 1/2$ the ASR lies exactly at the Fermi level: $E_R = E_F$. In the compounds CeAl_3 and CeCu_2Si_2 , the sixfold degeneracy of the cerium level ($j = 5/2, \nu = 6$) is split by the crystal field Δ_{CF} into three doublets. However, since in this case $T_K \ll \Delta_{\text{CF}}$, for low enough temperatures only the lowest doublet is effective, i.e., the compounds CeAl_3 and CeCu_2Si_2 behave like NKLs with " $j = 1/2$,"^{2,16,17} in which the ASR is located exactly at the Fermi level $-E_R = E_F$. It is this circumstance which gives rise to the low-temperature 10^2-10^3 -fold enhancement of the density of states at the Fermi level for such NKLs as CeAl_3 , CeCu_2Si_2 , UBe_{13} .

As the temperature is raised, the thermal spin fluctuation time τ_T decreases, and becomes smaller than the characteristic spin fluctuation time: $\tau_T \ll \tau_{\text{sf}}$, i.e., the thermal scattering suppresses the Kondo fluctuations. As a result of this, the ASR amplitude decreases as the temperature increases, and ultimately for $T \gg T_K$ the resonance disappears entirely.

The review is laid out in the following fashion: Section 2 contains an analysis of the properties of the substitutional solid solutions $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$, $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$ and $\text{Ce}_x\text{La}_{1-x}\text{Cu}_6$, which span the whole range of intermediate states between the normal metals (LaCu_2Si_2 , LaAl_3 and LaCu_6), and NKLs (CeCu_2Si_2 , CeAl_3 and CeCu_6); this allows us to elucidate the genesis of the unusual properties of NKLs. In Section 3, a summary is given of the basic low-temperature anomalies of the NKLs CeCu_2Si_2 , CeAl_3 , UBe_{13} and CeCu_6 ; a comparison is made between the available experimental data and estimates based on a model of N_i noninteracting Kondo impurities, each of which makes its own independent contribution to the formation of a gigantic ASR at $E = E_F$. It is shown that with regard to the ensemble of their low-temperature properties, NKLs have no analogues, and in fact must be investigated as a fundamentally new class of metallic systems. The possibility of a transition to coherent Kondo fluctuations is investigated for $T \ll T_K$.

We analyze the dependence of the CKS properties on the ratio between the Kondo temperature T_K and the magnitude Δ_{CF} of the splitting of the magnetic f level in the crystal field. It is shown that the transition from $T_K \ll \Delta_{\text{CF}}$ (a " $j = 1/2$ " CKS with $E_R = E_F$) to $T_K \gg \Delta_{\text{CF}}$ ($j = j_{\text{tot}}$, $E_R \neq E_F$) leads to a shift of the ASR relative to E_F which can mimic the transition from the CKS to a regime with variable valence. The contribution of effects due to a type of "Kondo collapse"^{50,51} are discussed with regard to their role in limiting the valence $\nu(\text{Ce})$ in cerium compounds. In Section 4, the exotic superconducting properties of the NKLs CeCu_2Si_2 and UBe_{13} are investigated. In conclusion we will mention experimental investigations which could advance the progress of understanding the nature of the unusual properties of NKLs and other HFSs.

2. THE KONDO-IMPURITY TO KONDO-LATTICE TRANSITION

2.1. Isostructural substitutional solid solutions in which a transition occurs from normal metal to Kondo lattice by way of a Kondo-impurity regime

In order to understand the nature of the low-temperature anomalies of nonmagnetic Kondo lattices, it is of great value to undertake the complex investigation of properties of isostructural substitutional solid solutions. In these systems, it is possible to induce a continuous progression through the entire range of intermediate states between normal metals and nonmagnetic Kondo lattices, including the rather well-understood Kondo-impurity regime. In this review, the behavior of NKLs will be investigated based on the sample compounds CeCu_2Si_2 and CeAl_3 , in which the localized magnetic moment is related to the $4f^1$ configuration of the Ce^{3+} ions.

Since lanthanum, which is located to the left of cerium in the periodic table, has almost the same ionic radius as cerium while its $4f$ shell is empty, the compounds LaCu_2Si_2 and LaAl_3 are normal nonmagnetic isomorphous homologues of the NKLs CeCu_2Si_2 and CeAl_3 . The compounds RE_2X_2 ($\text{RE} = \text{rare earth elements}$, $\text{X} = \text{silicon or germanium}$ and $\text{M} = \text{Fe, Co, Ni, Cu, Ag, ...}$) crystallize in the ThCr_2Si_2 type^{52,199} of structure (a centrosymmetric tetragonal structure) with intermediate layers of atoms perpendicular to the fourfold axis in the sequence RE-X-M-X-RE-... . The lattice parameters of CeCu_2Si_2 and LaCu_2Si_2 are extremely close^{53,54} (Table I), so that isostructural substitutional solid solutions can be made over the entire interval of composition $0 < x < 1$. An analogous situation obtains also for the compound CeAl_3 , which like LaAl_3 crystallizes in a hexagonal structure of the type Ni_3Sn (Table I). The unit cell volumes of CeCu_2Si_2 and CeAl_3 are smaller than the corresponding volumes for LaCu_2Si_2 and LaAl_3 . As a result of this, increasing the cerium content in $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ and $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$ leads to the appearance of a type of "chemical compression" effect.

Recently, studies have been carried out, based on the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ and $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$, of the nature of variations in the electrical,^{48,49,54,56} magnetic^{54,56,57} and thermoelectric^{56,58} properties, in the temperature depen-

TABLE I. Lattice parameters of the NKLs CeCu_2Si_2 , CeAl_3 , CeCu_6 and their normal homologues LaCu_2Si_2 , LaAl_3 , LaCu_6 .

Compound	Structure	a, Å	c, Å	v, Å ³
CeCu_2Si_2 ⁵³	Tetragonal	4.103 ± 0.005	9.94 ± 0.006	167.3
LaCu_2Si_2 ⁵⁹	»	4.147 ± 0.002	9.9137 ± 0.003	170.5
CeAl_3 ⁶⁵	Hexagonal	6.5506 ± 0.001	4.6092 ± 0.0008	513.9
LaAl_3 ⁵⁵	»	6.6603 ± 0.001	4.6173 ± 0.0008	532.1

Compound	Structure	a, Å	b, Å	c, Å
CeCu_6 ⁶⁴	Orthorhombic	8.144	5.091	10.130
LaCu_6 ⁶⁴	Monoclinic ($\beta = 91,49^\circ$)	5.156	10.238	8.168

dences of the Hall coefficients,^{54,56,59-61} and in the microcontact spectra,⁶² for the transition from normal metals (LaCu_2Si_2 and LaAl_3) through the Kondo-impurity regime ($x \ll 1$) to the concentrated Kondo systems (CKS) ($x \approx 1$), including the NKLs CeCu_2Si_2 and CeAl_3 . These data make it possible to trace step by step how the smooth variation in composition leads for $x \rightarrow 1$ to the appearance of a number of anomalies in these CKSs.

Analogous data have also been obtained for the solid solutions $\text{Ce}_x\text{La}_{1-x}\text{Cu}_6$ ($0 \leq x \leq 1$).⁶³⁻⁶⁶ In this series, LaCu_6 crystallizes in a monoclinic lattice where CeCu_6 has an orthorhombic structure; the monoclinic distortion of the orthorhombic structure for $x \leq 0.38$ is accompanied by a change in the angle $\gamma = 90^\circ$. If this distortion is neglected, one may assume that in the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_6$, as in $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$ and $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$, there is a progression over the entire range of compositions from a normal metal ($x = 0$) to an NKL ($x = 1$) based on the same crystal structure, with a unit cell whose volume decreases as $x \rightarrow 1$.

2.2. Variation of the electrical properties of the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ and $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$ as the composition is varied from $x \ll 1$ to $x \approx 1$

Substituting Ce for La in the normal nonmagnetic metal LaCu_2Si_2 leads to the appearance of distinct regions of logarithmic growth in the resistivity $\rho(T)$ as the temperature falls in the region $T \lesssim 60$ K (Fig. 1). For concentrations $x \leq 0.3$, beyond the logarithmic regions of $\rho(T)$, saturation sets in, leading eventually to a plateau.^{54,56} Such dependences of $\rho(T)$ are characteristic of the behavior of Kondo impurities in a normal metal.^{23,29} A further increase in x in the alloy $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ leads to a change in the form of the $\rho(T)$ curves and the appearance of a maximum in the function $\rho(T)$ for $T = T_{\text{max}}^{(1)}$. The temperature $T_{\text{max}}^{(1)}$ increases with the cerium concentration (see Fig. 1) at an average rate $dT_{\text{max}}^{(1)}/dx \approx 0.11$ atomic % Ce. The maximum of $\rho(T)$ for $T = T_{\text{max}}^{(1)}$ is not connected with the magnetic transition to a frozen-spin state, since the variation in the magnetic susceptibility (see below) follows the usual paramagnetic variation $\chi(T)$ near $T = T_{\text{max}}^{(1)}$.

From the data on $\rho(T)$ (see Fig. 1) we can estimate the concentration dependence of the Kondo temperature

$T_K(x)$. However, determining T_K from the curves of $\rho(T)$ is, strictly speaking, correct only for the Kondo-impurity regime. However, we can make use of the following procedure to determine approximately the value of T_K in the CKS $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$. Extracting from the curves of $\rho(T)$ for $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ the "normal" phonon contribution to ρ , which is assumed to be equal to its value in $\rho(T)$ for LaCu_2Si_2 , and normalizing the difference $\Delta\rho$ so obtained to the cerium concentration, we can determine the "magnetic" contribution $\Delta\rho_m(T)$ to the resistivity. Let us now plot this data using the coordinates $\Delta\rho_m = f(\ln(T/T_K))$, and use T_K as a fitting parameter. We then arrange that the curves $\Delta\rho_m + f(\ln(T/T_K))$ for different x are described as well as possible by a single functional dependence on the coordi-

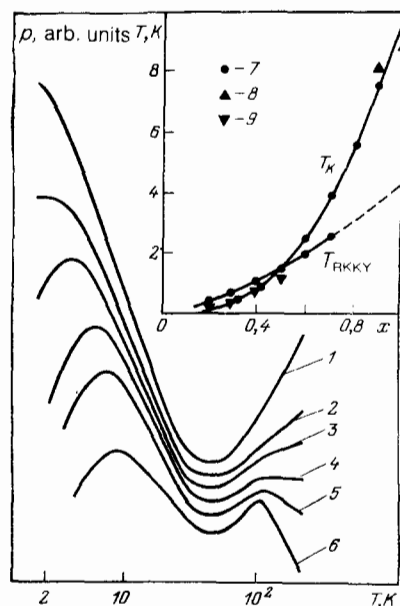


FIG. 1. Temperature dependence of the resistivity of $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ for the compositions $x = 0.2$ (1), 0.4 (2), 0.5 (3), 0.7 (4), 0.9 (5) and 1.0 (6).^{54,56} The inset shows the concentration dependences of the two characteristic temperatures T_K and T_{RKKY} for the same alloys. The quantity T_K is determined from data on $\rho(T)$ (7), using the minimum of the negative magnetoresistance (8) and from data on the magnetic susceptibility $T_K = \theta/4$ (9).

nates. This procedure gives the dependence of T_K on x , which is shown in the inset of Fig. 1. Analogous results for $T_K(x)$ can also be obtained by deriving T_K from the minimum of the negative magnetoresistance (NMR): $T_K = T_{\min}^{\text{NMR}} \times 2$.⁶⁸

In the alloys $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$ and $\text{Ce}_x\text{La}_{1-x}\text{Cu}_6$ an analogous transformation occurs in the dependence of $\rho(T)$ as the cerium content is increased.^{60,61,63,64} However, in the case of $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$ the low-temperature maximum of $\rho(T)$ is observed against a background high-temperature maximum of ρ for $T = T_{\max}^{(2)}$, whereas for $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ the temperatures $T_{\max}^{(1)}$ and $T_{\max}^{(2)}$ differ by more than an order of magnitude, and the maxima of $\rho(T)$ for $T = T_{\max}^{(1)}$ and $T = T_{\max}^{(2)}$ are not superimposed on one another: $T_{\max}^{(2)}(\text{CeCu}_2\text{Si}_2) \approx 110 \text{ K}$,^{54,56} $T_{\max}^{(2)}(\text{CeAl}_3) \approx 40 \text{ K}$.^{60,61,69} The high-temperature maximum in $\rho(T)$ for $T = T_{\max}^{(2)}$ in $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ and $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$ is associated with inelastic Kondo scattering by a doublet located above the ground-state doublet, since in the crystal fields of CeCu_2Si_2 and CeAl_3 the sixfold degeneracy of the Ce^{3+} level ($j = 5/2$) is split into three doublets: at 0°K , $\Delta_{\text{CF}_1} = 140^\circ\text{K}$ and $\Delta_{\text{CF}_2} = 364^\circ\text{K}$ in CeCu_2Si_2 ⁷⁰ and $\Delta_{\text{CF}_1} = 50^\circ\text{K}$, $\Delta_{\text{CF}_2} = 114^\circ\text{K}$ in CeAl_3 .⁷¹ The energy intervals between the lowest doublets (140°K in CeCu_2Si_2 and 50°K in CeAl_3) roughly correspond to the temperatures $T_{\max}^{(2)}$ in these compounds. In the temperature range $T > \Delta_{\text{CF}_1}$, Δ_{CF_2} , using the theory of Hanzawa, Yamada and Yosida⁷² we can determine the Kondo temperature T_K^h , taking into account the scattering by the upper doublets, in the following form:

$$T_K^h = (T_K \Delta_{\text{CF}_1} \Delta_{\text{CF}_2})^{1/3} \quad (2.1)$$

The temperatures T_K^h found from the known T_K , Δ_{CF_1} and Δ_{CF_2} are 32°K and 80°K for CeAl_3 and CeCu_2Si_2 respectively. The values of T_K^h thus obtained are in reasonable agreement with the temperatures $T_{\max}^{(2)}$ in these compounds.

In $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ and $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$, the increase in the Kondo temperatures as $x \rightarrow 1$ can be related either to the change in the volume of the unit cell ("chemical compression") or to variation in the concentration of cerium ions. So as to separate out the contributions from these two factors, it is interesting to appeal to the data of Groft and Levine,⁵⁷ who investigated the dependence of T_K on x for the solid solutions $\text{Ce}_x\text{Sc}_{1-x}\text{Al}_2$ and $\text{Ce}_x\text{Y}_{1-x}\text{Al}_2$, in which an increase in the cerium concentration occurs against a background growth in the volume V of the unit cell ($V(\text{CeAl}_2) > V(\text{YAl}_2), V(\text{ScAl}_2)$), and leads to a decrease in T_K . These data attest to the fact that the growth in T_K is caused by "chemical compression" independently of the increase or decrease in the concentration of magnetic centers, i.e., for both $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ and $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$ the growth in T_K as $x \rightarrow 1$ (see Fig. 1) is apparently connected with the change in unit cell volume in going from LaCu_2Si_2 , LaAl_3 to CeCu_2Si_2 and CeAl_3 (see Table I). The conclusion that changes in unit cell volume play a determining role for the alloys investigated is confirmed by the noticeable rise in T_K under compression (see below).

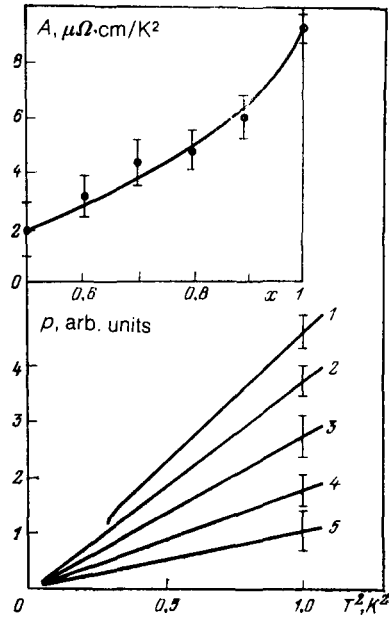


FIG. 2. Temperature dependences $\Delta\rho = AT^2$ for the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ for various $x = 1.0$ (1), 0.9 (2), 0.8 (3), 0.6 (4) and 0.5 (5).⁵⁶ The inset above shows the concentration dependence of the coefficient A .

For low temperatures $T < T_{\max}^{(1)}$ in the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ for $x \geq 0.4$ we observe a fall in the resistance which follows a quadratic law: $\Delta\rho = AT^2$ (Fig. 2). The coefficient A substantially increases as $x \rightarrow 1$ and reaches a value of $\sim 10 \mu\Omega\text{-cm}/^\circ\text{K}^2$ in CeCu_2Si_2 ^{8,17,56} and $\sim 35 \mu\Omega\text{-cm}/^\circ\text{K}^2$ in CeAl_3 .⁶ In single crystal CeCu_6 for temperatures $T < T_{\max}^{(1)}$ the law $\Delta\rho = AT^n$ is also obeyed; however, the values of the parameters A and n show significant anisotropy⁶³: for the orientation $i||a$ (i is the current), $A = 26.3 \mu\Omega\text{-cm}/^\circ\text{K}^n$ and $n = 2.01$; for $i||b$, $A = 13.8 \mu\Omega\text{-cm}/^\circ\text{K}^n$ and $n = 1.71$; for $i||c$, $A = 39.3 \mu\Omega\text{-cm}/^\circ\text{K}^n$, $n = 2.07$. It is natural to relate the temperature variation $\Delta\rho = AT^2$ in CKS to electron-electron scattering, for which

$$AT^2 = \rho(T) - \rho(T \rightarrow 0) = \frac{\pi^2 e^2 m_0 \cdot 76,06}{16n h^3} \left(\frac{T}{T_F^*} \right)^2; \quad (2.2)$$

here, m_0 is the free-electron mass, n the density of sd -electron states, and T_F^* is the degeneracy temperature.

2.3. Variation of the magnetic properties of the alloy $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ in the transition Kondo-impurity to Kondo-lattice

The temperature dependence of the differential magnetic susceptibility $\chi(T)$ in the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ for cerium contents $0.2 < x < 0.7$ is characterized by monotonic growth in χ as the temperature falls, down to $T = T_{\max}^x$ at which a kink is observed in the $\chi(T)$ curve (Fig. 3), corresponding to the transition from a paramagnetic material to a state with frozen-in spins (either an antiferromagnetic material or a spin glass).^{54,57} At temperatures above the magnetic

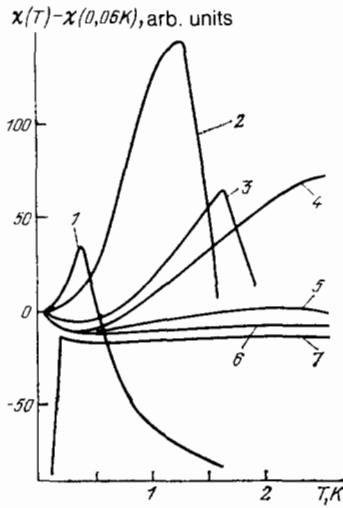


FIG. 3. Temperature dependences of the differential magnetic susceptibility $\chi(T) - \chi(0.06 \text{ K})$ for the alloys $\text{Ce}_x \text{La}_{1-x} \text{Cu}_2 \text{Si}_2$ for different $x = 0.2$ (1), 0.5 (2), 0.6 (3), 0.7 (4) and 0.9 (5).⁵⁴ Curve 6 is single-crystal $\text{CeCu}_2 \text{Si}_2$; curve 7 corresponds to $\chi(T)$ for polycrystalline $\text{CeCu}_2 \text{Si}_2$.

transition T_{max}^{χ} , the dependence of $\chi(T)$ is described by the Curie-Weiss law $\chi(T) \sim C/(T + \theta)$ with constant $\theta < 0$. From the value of θ we can estimate the Kondo temperature, taking advantage of the relation obtained by Gruner and Zawadowsky: $T_K \approx |\theta|/4$. Both estimates of the concentration dependences of the Kondo temperature—from $\rho(T)$ and from $\chi(T)$ (see the inset of Fig. 1)—show that the passage from $x \ll 1$ to $x \approx 1$ in $\text{Ce}_x \text{La}_{1-x} \text{Cu}_2 \text{Si}_2$ leads to a substantial increase in the temperature $T_K(x)$.^{17,54,56}

In the range $0.7 \leq x \leq 1.0$, in the alloys $\text{Ce}_x \text{La}_{1-x} \text{Cu}_2 \text{Si}_2$ there has been observed^{54,56} an unusual variation in the magnetic properties as the concentration of magnetic ions is varied (see Fig. 3): starting at $x = 0.7$ to 0.8, increasing the concentration of the magnetic component leads not to an enhancement of the tendency to transform to a state with frozen-in spins, but on the contrary to an effective suppression of this tendency. For $x \geq 0.8$, the kink in $\chi(T)$ for $T = T_{\text{max}}^{\chi}$ begins to smooth out, and the temperature T_{max}^{χ} itself decreases. For $x \rightarrow 1$ there also occurs a significant weakening in the temperature dependence of the paramagnetic susceptibility $\chi(T)$, which for alloys with $x \approx 1$ gives rise to a temperature-independent enhancement of the Pauli paramagnetism. Finally, in the limiting compound $\text{CeCu}_2 \text{Si}_2$ with maximum content of Ce^{3+} magnetic ions for the series $\text{Ce}_x \text{La}_{1-x} \text{Cu}_2 \text{Si}_2$, in place of a magnetic transition we observe a transition to the superconducting state, which is signaled by the presence of a diamagnetic anomaly in χ at $T = T_c$ (see Fig. 3). The absence of a magnetic transition in $\text{CeCu}_2 \text{Si}_2$ is also established in experiments using neutron scattering⁶⁶: neither a change in the amplitude of the Bragg peaks nor the appearance of additional lines are observed for temperatures down to $T = 0.02 \text{ K}$.

In normal metals, including $\text{LaCu}_2 \text{Si}_2$, the magnetic ions interact with each other in an indirect fashion via

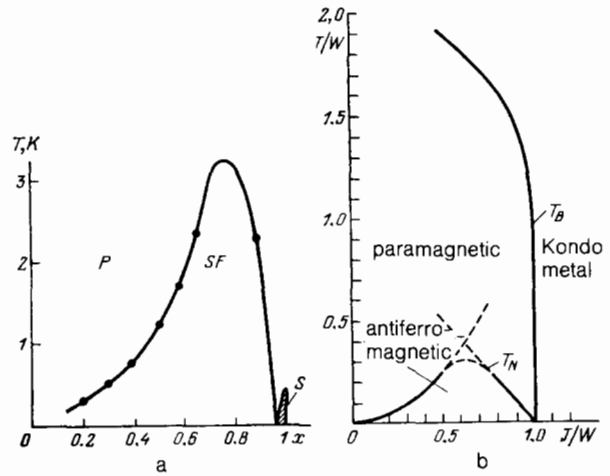


FIG. 4. (a) Phase diagram of the magnetic properties of the alloys $\text{Ce}_x \text{La}_{1-x} \text{Cu}_2 \text{Si}_2$; P—paramagnetic, SF—frozen-in spin state, antiferromagnet or spin glass, S—superconducting phase; (b) Phase diagram of magnetic properties of Kondo lattices investigated by Doniach⁷⁴ within the framework of a one-dimensional model of Kondo “necklace” type; J/W —exchange interaction constant normalized to the width of the conduction band.

the Ruderman-Kittel-Kasuya-Yosida spin-density oscillations.³⁸⁻⁴⁰ Assuming that in the alloys $\text{Ce}_x \text{La}_{1-x} \text{Cu}_2 \text{Si}_2$ the RKKY mechanism corresponds to a transition below T_{max}^{χ} to a state with frozen-in spins, we can determine a characteristic temperature $T_{\text{RKKY}}(x)$ from the value T_{max}^{χ} in the following way.^{17,56} For those concentrations x ($0.2 \leq x < 0.7$), for which the quantity $T_{\text{max}}^{\chi}(x)$ is a monotonic function of x , let us set $T_{\text{RKKY}}(x) = T_{\text{max}}^{\chi}(x)$ (Fig. 4). Outside of the interval, the curve $T_{\text{RKKY}}(x)$ can be drawn using an extrapolation of $T_{\text{RKKY}}(x)$ into the region $0.7 < x < 1.0$ (the dotted line in the inset of Fig. 1).

The intensity of the RKKY interaction in metals depends on the exchange interaction parameter J between f and sd electrons, and the spacings R_{ij} between the i th and j th magnetic ions³⁸⁻⁴⁰:

$$T_{\text{RKKY}}(x) \sim \frac{J^2(x)}{R_{ij}^3} \sim J^2(x) x. \quad (2.3)$$

Comparing this expression with the available experimental data (see Fig. 4) in the range $x = 0.2-0.7$, we can conclude that the more-than-linear growth of $T_{\text{RKKY}}(x)$ is related to an increase in the parameter J for increasing x . The dependence $T_K(x)$ also points to an increase in J (see Fig. 1), since according to (1.1) the Kondo temperature grows exponentially with increasing J .

Expression (2.3) does not explain the decrease in T_{max}^{χ} in the region $x = 0.7-1.0$, since it is obtained under conditions of constancy of the effective magnetic moment $\mu_{\text{eff}}(\text{Ce}^{3+})$. Consequently, the points in the inset to Fig. 1 correspond only to the expected variation of T_{RKKY} under the condition that $\mu_{\text{eff}}(\text{Ce}^{3+}) = \text{const}$. The experimentally-observed decrease of $T_{\text{max}}^{\chi}(x)$ (see Fig. 1) attests to the effective low-temperature suppression of the magnetic moment $\mu_{\text{eff}}(\text{Ce}^{3+})$ as $x \rightarrow 1$ in the $\text{Ce}_x \text{La}_{1-x} \text{Cu}_2 \text{Si}_2$ series.

Once the characteristics of the variation of the two parameters T_K and T_{RKKY} (see Fig. 1) are known, it is practical to carry out an analysis of the $T-x$ phase diagram (see Fig. 4a) for the alloys $Ce_x La_{1-x} Cu_2 Si_2$ within the framework of the approach assumed by Doniach,⁷⁴ who in 1977 predicted theoretically for the first time the non-monotonic dependence of the magnetic transition temperature on the exchange interaction parameter J (Fig. 4b), based on a one-dimensional "Kondo necklace" model. For the alloys $Ce_x La_{1-x} Cu_2 Si_2$, because of a kind of "chemical compression" effect the quantity J turns out to be a monotonically increasing function of the cerium concentration x . Therefore, the change in magnetic properties as we go from $x \ll 1$ to $x \approx 1$ is naturally related to the crossover regime: magnetic for $T_K \ll T_{RKKY}$ to nonmagnetic for $T_K \gg T_{RKKY}$ (see Fig. 1). The crossover regime is caused by the different dependences of T_{RKKY} and T_K on the parameter J . The indirect exchange interaction temperature is a *quadratic* function of J at the same time that the effectiveness of the Kondo compensation of the magnetic ions depends *exponentially* on J (1.1). Therefore, in systems with a sufficiently large parameter J , the exponential dependence of $T_K(J)$ will "overtake" the power-law dependence $T_{RKKY}(J)$ and bring about the regime $T_K \gg T_{RKKY}$. From this point of view, for $T \rightarrow 0$ the nonmagnetic state in alloys $Ce_x La_{1-x} Cu_2 Si_2$ with $x \approx 1$ appears because of effective suppression of the magnetic moments of the Ce^{3+} ions by the Kondo processes, which for $x \rightarrow 1$ ensure that the inequality $T_K \gg T_{RKKY}$ holds.

The $T-x$ phase diagram (see Fig. 4) for $Ce_x La_{1-x} Cu_2 Si_2$ confirms the theoretical predictions of Doniach.⁷⁴ Earlier,^{75,76} a nonmonotonic variation in the magnetic transition temperature T was also observed in the compound CeAg when J was varied by applying external pressure.

It is interesting to note that the boundary compound $CeCu_2 Si_2$ which in Doniach's terminology is a "Kondo metal," not only does not convert into a magnetic state but in addition is found to be an unusual superconductor (see below).

The dependence of the temperature T_M on the normalized exchange interaction constant J/W (W is the band width), taking into account the difference in behavior of $T_K(J)$ and $T_{RKKY}(J)$ [see (2.3) and (1.1)], can be used to classify the various types of CKS. Compounds with $T_K \gg T_{RKKY}$ and $T_M = 0$ are commonly called *nonmagnetic* CKSs. Nonmagnetic CKSs with a periodic arrangement of Kondo centers have been given the name "nonmagnetic Kondo lattices," e.g., the compounds $CeCu_2 Si_2$, $CeAl_3$, $CeCu_6$, UBe_{13} (see below). The intermediate situation with $T_K \approx T_{RKKY}$ but with a non-zero magnetic transition temperature $T_M \neq 0$ corresponds to a *magnetic* CKS. A subset of these are the magnetic Kondo lattices: CeB_6 , $CeAl_2$, $CeIn_3$, etc. In these compounds, decreasing the temperature leads to the appearance of obvious Kondo anomalies. Nevertheless, the transition to a state with frozen-in spins makes the formation of a nonmagnetic Kondo state impossible. Metallic compounds based on rare-earth elements, in which the indirect exchange interaction dominates the Kondo spin

fluctuations and $T_{RKKY} \gg T_K$ (J small) are found to be normal magnetic $4f$ metals. In these compounds, the low-temperature Kondo anomalies are completely absent, since for higher temperatures the magnetic transition occurs, the spins are locally frozen-in and Kondo spin fluctuations for $T < T_{RKKY}$ are impossible.

2.4. Low-temperature anomalies in the Hall effect of concentrated Kondo systems

In passing from a normal metal ($x = 0$) to a Kondo lattice ($x = 1$), the temperature dependence of the Hall coefficient $R_H(T)$ in the alloys $Ce_x La_{1-x} Cu_2 Si_2$, $Ce_x La_{1-x} Al_3$ and $Ce_x La_{1-x} Cu_6$ changes in the following way^{54,56,59-61,63-66}: in the normal metals $LaCu_2 Si_2$, $LaAl_3$ and $LaCu_6$, a metallic variation of $\rho(T)$ is observed against the background of the temperature-independent Hall coefficient $R_H(T)$ (Fig. 5). In the Kondo-impurity regime, for $Ce_x La_{1-x} Cu_2 Si_2$ and $Ce_x La_{1-x} Al_3$ an increase in $R_H(T)$ by a factor of 1.5 to 2 occurs as the temperature goes from room temperature to liquid helium. A qualitatively similar behavior of $R_H(T)$ was observed earlier in $Au(Fe)$ ²⁵ and $La(Ce)$.⁷⁷

The passage from $x \ll 1$ to $x \approx 1$ in the alloys $Ce_x La_{1-x} Cu_2 Si_2$, $Ce_x La_{1-x} Al_3$, $Ce_x La_{1-x} Cu_6$ leads to a significant enhancement in the low-temperature anomalous Hall coefficient: the ratio $R_H(4.2\text{ K})/R_H(70\text{ K})$, which characterizes the growth of $R_H(T)$ attains the value 12-15 for $x \rightarrow 1$ (see inset to Fig. 5). It must also be noted that the magnitude of this ratio depends on the cerium concentration in a nonlinear way—the most significant enhancement in the nonlinear growth of $R_H(T)$ occurs in the interval $x = 0.7-1.0$.

The Hall anomalies appear most sharply in NKLs: whereas in a normal metal a drop in temperature leads to a decrease in the resistivity and in practice does not influence

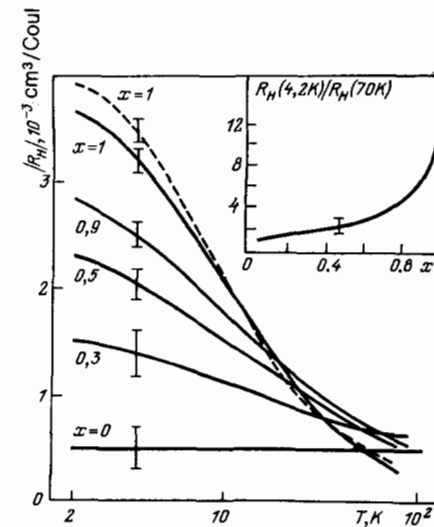


FIG. 5. Temperature dependences of the Hall coefficient $R_H(T)$ for $Ce_x La_{1-x} Cu_2 Si_2$ for various x .^{54,56} Continuous curve—data for $CeCu_2 Si_2$. In the inset is shown the concentration dependence of the ratio $R_H(4.2\text{ K})/R_H(70\text{ K})$.

the Hall coefficient, in the NKLs CeAl_3 , CeCu_2Si_2 and CeCu_6 , on the contrary, the resistance changes by as much as a factor of 1.5, while the Hall coefficient increases by 15–20 times in the temperature interval $T = 2\text{--}100\text{ K}$.

The sign of the Hall coefficient $R_H(4.2\text{K})$ in the NKLs CeAl_3 , CeCu_2Si_2 and CeCu_6 is positive. Measurement of the polarity dependence of the Hall voltage U_H in static fields up to $H = 120\text{ kOe}$ shows that the dependence $U_H(H)$ is linear up to $H = 90\text{ kOe}$. There is also a certain tendency to saturate at $U_H(H, T)$ for $H \geq 100\text{ kOe}$ for $T_0 = 1.5\text{ K}$. For temperatures higher than T_0 , this tendency manifests itself to a lesser degree.

For IVCs unusual behavior of $R_H(T)$ was observed in Ref. 78 for the compound CePd_3 , where $R_H(T = 100\text{ K}) \approx 2.5 \times 10^{-3}\text{ cm}^3/\text{C}$; decreasing the temperature induces a sign change in the Hall coefficient $R_H(T = 4.2\text{ K}) = -1.0 \times 10^{-3}\text{ cm}^3/\text{C}$. Among the compounds of type RECu_2Si_2 ($\text{RE} = \text{rare-earth element}$) investigated in Ref. 79— YCu_2Si_2 (a nonmagnetic normal metal), GdCu_2Si_2 (an antiferromagnet with $T_N = 13.5\text{ K}$), TbCu_2Si_2 (an antiferromagnet with $T_N = 11.5\text{ K}$) and CeCu_2Si_2 —the amplitude of the low-temperature growth in the Hall coefficient of CeCu_2Si_2 was significantly larger than even that of isostructural metals with magnetic transitions.

Among the NKLs based on $5f$ elements, a low-temperature growth in $R_H(T)$ was observed in the compound UBe_{13} .⁸⁰ For UBe_{13} , just as for CeCu_2Si_2 , CeAl_3 and CeCu_6 , $R_H(4.2\text{ K}) > 0$, while the dependence $U_H(H)$ goes to saturation for a magnetic field $H \sim 90\text{ kOe}$.

2.5. Thermoelectric properties of the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ and $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$

The first measurements of thermoelectric properties of solid solutions in which the transition from Kondo impurity to Kondo lattice was induced were made by Van Aken and coauthors⁶⁹ on the alloys $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$ ($x = 0; 0.01; 0.1; 0.5; 1.0$). Analogous results were obtained later on for the alloys $\text{Ce}_x\text{La}_{1-x}$,⁸¹ $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ ^{56,59} and $\text{Ce}_x\text{La}_{1-x}\text{Cu}_6$.⁶⁶ The temperature dependence of the Seebeck coefficient $S(T)$ for $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ is shown in Fig. 6. The curves $S(T)$ are characterized by the presence of two distinctive features: a maximum for $T = T_{\text{max}}^{(2)} \approx 150\text{ K}$ and an extremum for $T = T_{\text{extr}}^{(1)} \approx 10\text{ K}$. Whereas the values of these temperatures are almost constant over the entire interval of concentration ($x = 0\text{--}1.0$), the low-temperature features themselves are significantly altered in going from $x \ll 1$ to $x = 1$. First of all (see Fig. 6), increasing the cerium content leads to a growth in the amplitude of the low-temperature maximum. Subsequently, for $x \rightarrow 1$, $S(T = T_{\text{extr}}^{(1)})$ changes sign, and the absolute value of $S(T)$ at the minimum rapidly increases with cerium content up to $|S(T_{\text{extr}}^{(1)})| \approx 10\text{ }\mu\text{V}/\text{K}$ for CeCu_2Si_2 .⁸

In order to analyze the way the temperature dependence of the thermoelectric power varies for the series $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$, it is necessary to include the following factors: it is well-known that in Kondo systems the presence of the low-temperature maximum in $S(T)$ is an indication of impurity-impurity interactions, while an isolated Kondo im-

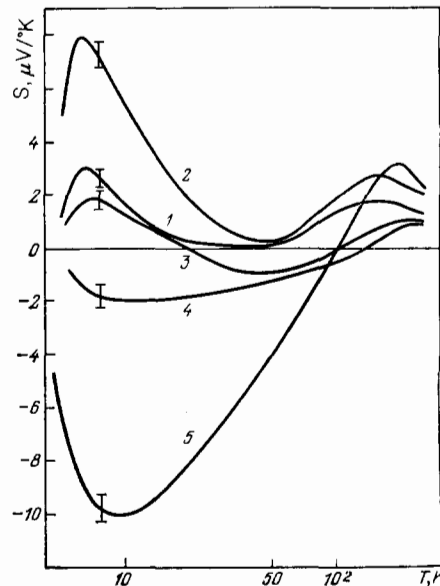


FIG. 6. Temperature dependences of the Seebeck coefficient $S(T)$ in the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ for various cerium contents $x = 0.2$ (1), 0.3 (2), 0.7 (3), 0.9 (4).⁵⁶ The curve 5 is single-crystal CeCu_2Si_2 .

purity gives a negative contribution to $S(T)$; in the region $T < T_K$, this contribution in first approximation should not depend either on temperature or on the concentration of Kondo-centers.⁸³ Consequently, we can suppose that the maximum in $S(T)$ for $T_{\text{extr}}^{(1)} \sim 10\text{ K}$ (see Fig. 6) reflects a tendency of the magnetic Ce^{3+} ions to pass to a state with frozen-in spins. The maximum amplitude of the feature $S(T = T_{\text{extr}}^{(1)})$ corresponds to that concentration $x = 0.3\text{--}0.4$ for which the sharpest kink appears in the temperature dependence of the magnetic susceptibility (see Fig. 3). The suppression of the tendency toward magnetic ordering for $x \geq 0.7$ (see Figs. 3, 4) is accompanied by a change in the sign of S and rapid growth in the absolute value of $S(T)$ at the minimum. For $T < T_{\text{extr}}^{(1)}$ in CeCu_2Si_2 the dependence of $S(T)$ is found to be linear:⁸

$$S(T) = - \left(\frac{\pi^2}{3} \frac{k_B}{e} \frac{1}{T_F^*} \right) T = -aT. \quad (2.4)$$

The linearity of $S(T)$ is usually interpreted as a result of Fermi-liquid effects, since for an isolated Kondo impurity the value of S should not depend on T for $T \ll T_K$.^{83,84}

For the NKL CeCu_2Si_2 the coefficient a [in (2.4)] is anomalously large: $a \sim 7\text{--}9\text{ }\mu\text{V}/\text{K}^2$. The effective temperature T_F^* for such a value of a is $10^{\circ}\text{--}30^{\circ}\text{ K}$. This result is found to be in agreement with the value of T_F^* found from the Fermi-liquid behavior of the resistivity: $\Delta\rho = AT^2$ (see Fig. 2). We note that the effective temperature $T_F^* \sim 10\text{ K}$ is astonishingly far below that of a metal with carrier concentration $n \sim 10^{22}\text{ cm}^{-3}$; in normal metals, the corresponding temperature is $T_F^* \sim 10^4\text{--}10^5\text{ K}$.

In the NKLs CeCu_2Si_2 , CeAl_3 and CeCu_6 , the absolute values of the Seebeck coefficient are roughly two orders of magnitude higher than those of normal metals. Using the formula

$$S(T) = \frac{\pi k_B^2}{3|e|} T \left(\frac{1}{g(E)} \frac{dg(E)}{dE} - \frac{3}{2E} \right) \Big|_{E=E_F}, \quad (2.5)$$

the enhancement of $|S(T)|$ in NKLs can be viewed as a consequence of the existence of a narrow peak in the density of states near the Fermi level, since for low temperatures the value of $S(T)$ in (2.4) is determined by the logarithmic derivative of the density of states $g(E)$.

It must be pointed out that in formula (2.5) no account was taken of contributions to the thermoelectric power arising from odd (in energy) terms in the probability of scattering by magnetic impurities. Since the thermoelectric power is given in terms of an integral into which is inserted an odd function of the quasiparticle energy, the integral will not vanish in the case where this "oddness" is compensated by odd terms appearing either in the scattering probability or in the expansion of the density of states function.⁸³ Regrettably, a theory for CKSs which takes both these possibilities into account is to our knowledge not yet available.

The extremely low degeneracy temperature $T_F^* \sim 10^4 \text{ K}$ for NKLs corresponds to a very large fermion effective mass—of the order of $(10^2-10^3)m_0$. The contribution of such heavy quasiparticles to the low-temperature thermal conductivity is anomalously small.⁸ Thus, for CeCu_2Si_2 even for $T < 100 \text{ K}$ the electronic contribution K_e to the thermal conductivity is negligibly small compared to the phonon contribution K_{ph} . For normal superconductors the condition $K_e \ll K_{ph}$ is fulfilled below the critical temperature (i.e., for $T < T_c < 25 \text{ K}$), whereas for the NKL CeCu_2Si_2 the inequality $K_e \ll K_{ph}$ applies starting at a temperature $T \sim 100 \text{ K}$,⁸ far exceeding the value of T_c for all known superconductors.

2.6. Micro-contact spectroscopy of $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ alloys

An investigation of the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ by the method of micro-contact spectroscopy was carried out in Refs. 62 and 85, and for the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_6$ by Sato and coauthors.⁶⁶ In the normal metal LaCu_2Si_2 , a ballistic regime can be realized within the contact, in which the contact size d is much smaller than the inelastic mean free path l of electrons, and peculiarities are observed⁸⁵ in the second derivative of the I - V characteristic $d^2V/dI^2(V)$ of the micro-contact as a function of V ; these peculiarities are related to the electron-phonon interaction function $g(\omega)$. In the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$, because of scattering by the magnetic Ce^{3+} ions, the mean free path l is much reduced, and current flow takes place within the thermal regime ($l \ll d$). In the thermal regime, the dependence of the differential resistance $R_D = dV/dI$ on the voltage V (Fig. 7) is qualitatively similar to the dependence of the resistivity ρ on temperature (see Fig. 1), i.e., the curve $R_D(V)$ has two maxima, at 5 and 67 mV. For $V \rightarrow 0$, $R_D(V)$ is described by a quadratic function, while in the region $V = 7-30 \text{ mV}$ $R_D(V)$ decreases logarithmically (see the inset to Fig. 7). The correlation between $R_D(V)$ and $\rho(T)$, which is characteristic of the thermal regime, implies that the temperature T within the contact is related to that of the heat bath T_0 in the following way:

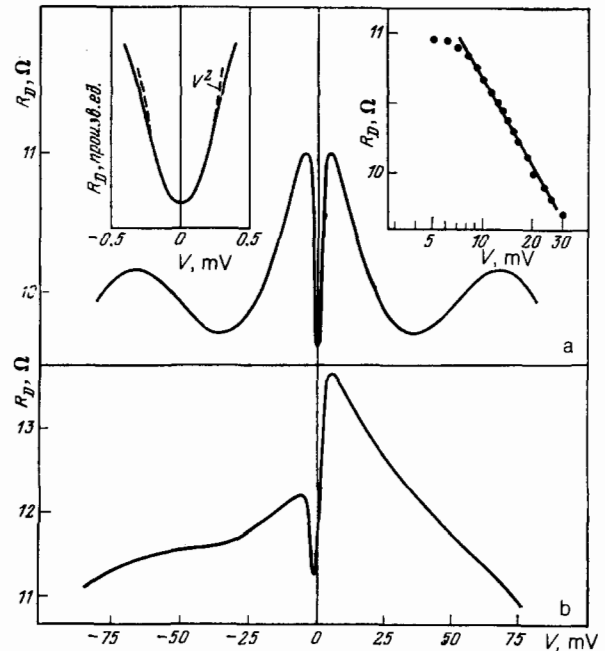


FIG. 7. Differential resistance function $R_D(V)$ of a homogeneous micro-contact made of CeCu_2Si_2 ($T_0 = 4.2 \text{ K}$) (a) and hetero-contact $\text{Cu-CeCu}_2\text{Si}_2$ (b) (for $V \rightarrow 0$, current flows from Cu to CeCu_2Si_2).⁶²

$$T^2 = T_0^2 + \frac{V^2}{4L}; \quad (2.6)$$

here, L is the Lorentz number. In the thermal regime, using the experimental curves for $R_D(V)$ we can calculate the dependence of the microcontact resistance on temperature, $R_{mc}^{\text{theor}}(T)$, using a linear relation between T and V for $T^2 \gg T_0^2$. In Ref. 62 it was shown that a good correspondence between the calculated curves $R_{mc}^{\text{theor}}(T)$ and the experimental curves $R_{mc}(T)$ is observed in the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ over a rather wide temperature range $T = 10^2-100 \text{ K}$, if we set $L/L_0 = 5$ in (2.6), where L is the standard value of the Lorentz number. Thus, there is a peculiar analogy between the microcontact spectra $R_D(V)$ for the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ (see Fig. 7) and the temperature dependences of the resistivity $\rho(T)$: in the regime $T \gg T_0$, the scale of temperature is proportional to the scale of voltage.

The microcontact spectra obtained for contact to homogeneous materials, including the $R_D(V)$ curves for $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ - $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ contacts are symmetric around the $V=0$ axis (see Fig. 7a).^{62,85} At the same time, for hetero-contacts of the type $\text{Cu-CeCu}_2\text{Si}_2$,^{62,85} or $\text{Mo-CeCu}_2\text{Si}_2$ and $\text{Pt-CeCu}_2\text{Si}_2$,⁸⁷ a significant asymmetry is observed in the curves $R_D(V)$ (see Fig. 7b), which can be characterized by introducing the odd part of the differential resistance $R_D^{\text{odd}}(V) = (1/2) \times (R_D^+(V) - R_D^-(V))$. For the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ the function $R_D^{\text{odd}}(V)$ is well correlated with the dependence of the thermoelectric power coefficient on temperature for bulk samples (Fig. 8a).^{56,58} Therefore, we can suppose that the asymmetry in the differential resistance of the microcontact (see Fig. 7b) is con-

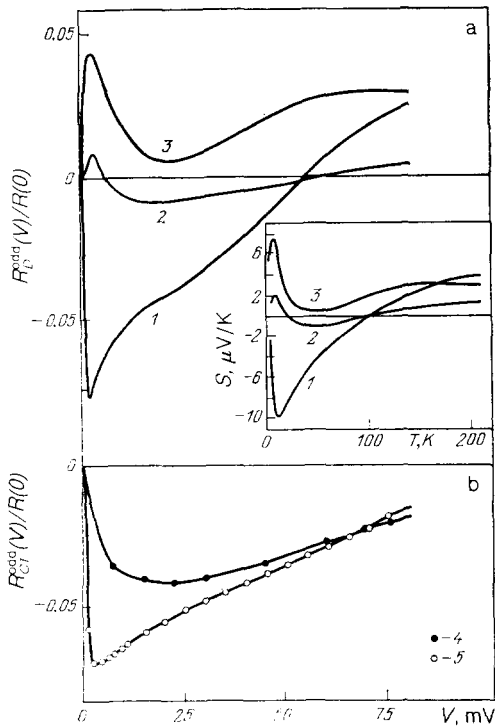


FIG. 8. (a) Experimental dependences of the asymmetric part of $R_D^{\text{odd}}(V)$ for hetero-contacts: Cu-CeCu₂Si₂ (1); Cu-Ce_{0.7}La_{0.3}Cu₂Si₂ (2); CuCe_{0.3}La_{0.7}Cu₂Si₂ (3). In the inset are shown the temperature dependences of the Seebeck coefficient $S(T)$ measured in Refs. 56 and 58 for bulk samples of the same Ce_xLa_{1-x}Cu₂Si₂ compounds $x = 1.0$ (1), 0.7 (2) and 0.3 (3). (b) Comparison of the theoretical dependence calculated using formula (2.7) for the hetero-contact CeCu₂Si₂-Cu (4) with experimental data (5).

nected with the appearance of the Seebeck and (or) Peltier effects.^{62,87,88} The contribution of the Seebeck and Peltier effects to $R_D^{\text{odd}}(V)$ can be estimated from the formulae

$$\frac{R_{\text{st}}^{\text{odd}}(V)}{R(0)} \approx \int_{T_0}^T [S_1(T) - S_2(T)] \frac{dT}{V} \quad (2.7)$$

$$R_{\text{st}}^{\text{odd}}(V) \approx \frac{\partial R_{\text{gr}}}{\partial T} [S_1(T) - S_2(T)] \frac{T_0}{V}, \quad (2.8)$$

where $R_{\text{st}}^{\text{odd}} = 1/2(R_{\text{st}}^+ - R_{\text{st}}^-)$; R_{st}^+ , R_{st}^- are the static microcontact resistances for the two polarities:

$$R_{\text{st}}(V) = \frac{V}{I(V)} = V^{-1} \int_0^V R_D^{\text{st}}(v) dv;$$

S_1 and S_2 are the thermoelectric powers of the contact materials. Estimates using formulae (2.7) and (2.8) show that the basic contribution to the asymmetry in the microcontact spectrum (see Fig. 7b) is due to the thermoelectric power, while the Peltier effect contribution is an order of magnitude smaller.⁶² Since we know the proportionality coefficient between voltage and temperature from a comparison of the curves for $\rho(T)$ and $R_{\text{mc}}^{\text{theor}}(V)$, and also the value of the thermoelectric power for bulk samples of CeCu₂Si₂,^{56,55} we can compare $R_{\text{st}}^{\text{odd}}$ calculated using formula (2.7) with the experimental results (Fig. 8b). This comparison shows^{62,85}

that in the region $V > 50$ mV there is good agreement between experiment and calculations using formula (2.7). However, as $V \sim T$ increases, the curves $(R_{\text{st}}^{\text{odd}}(V))_{\text{theor}}$ and $(R_{\text{st}}^{\text{odd}}(V))_{\text{exp}}$ begin to diverge. Therefore, we cannot explain the asymmetry of the curve $R_D(V)$ for heterocontacts of the type "normal metal-NKL" as being entirely due to the influence of thermoelectric effects. In the low-temperature region the additional contribution to the asymmetry in the differential resistance $R_D(V)$ is apparently due to significant differences in the electronic density of states $g(E_F)$ between the contacting materials. From this point of view, the asymmetry in $R_D(V)$ for heterocontacts of the type CuCeCu₂Si₂ (see Fig. 7b), which in the low voltage regime cannot be wholly reduced to contributions from thermoelectric effects, implies that the presence of asymmetry is due to the extremely narrow density of states peak at the Fermi level in CeCu₂Si₂.

In Ref. 66, the authors report a measurement of the asymmetry of the microcontact spectra and thermoelectric power of Ce_xLa_{1-x}Cu₆ alloys. Although in this case also there is significant agreement between the temperature dependence of the asymmetric part of the microcontact spectra and the thermoelectric power of bulk samples (this is especially striking as $x \rightarrow 1$, i.e., the NKL CeCu₆), the authors of Ref. 66 make no comment whatever on this circumstance. In our opinion, the situation in the case of Ce_xLa_{1-x}Cu₆ is analogous to that described above in Section 2 for Ce_xLa_{1-x}Cu₂Si₂.⁶²

Thus, the investigation of the galvanomagnetic, thermoelectric and magnetic properties of the isostructural solid solutions Ce_xLa_{1-x}Cu₂Si₂, Ce_xLa_{1-x}Al₃ and Ce_xLa_{1-x}Cu₆ allow us to establish the nature of the variation of the parameters $T_K(x)$, $T_{\text{RKKY}}(x)$, $A(x)$, $T_M(x)$, $R_H(4.2^\circ\text{K})/R_H(77^\circ\text{K})$ and $S_{\text{extr}}^{(1)}(x)$ as we go from the Kondo-impurity to the Kondo-lattice regime. This transition in Ce_xLa_{1-x}Cu₂Si₂ leads to a crossover regime—from magnetic ($T_K \ll T_{\text{RKKY}}$) to nonmagnetic ($T_K \gg T_{\text{RKKY}}$)—and is also accompanied by a regime of enhanced amplitude of Fermi-liquid effects. Experimental data on the system Ce_xLa_{1-x}Cu₆⁶³⁻⁶⁵ imply the possibility that a nonmagnetic ground state $T_K \gg T_{\text{RKKY}}$ is realized over the whole composition interval $0 < x \leq 1$ without any crossover regime. In these alloys, the crystal-field splitting is smaller than that of Ce_xLa_{1-x}Cu₂Si₂, and is estimated to be $\Delta_{\text{CF}} = 50-60^\circ\text{K}$; therefore the relation $T_K \gg T_{\text{RKKY}}$ in Ce_xLa_{1-x}Cu₆ can be fulfilled not only because of the increase of the exchange interaction J for $x \rightarrow 1$, but also (apparently), and principally, because of the presence of levels split off by the crystal field. As was shown in Refs. 72 and 90-93, even in the case $T_K \ll \Delta_{\text{CF}}$, the orbital degeneracy of the f states leads in CKSs to dominance of the local Kondo spin fluctuations over the RKKY interaction between magnetic centers; as the quantity $\nu = 2j + 1$ grows in comparison with $j = 1/2$, Kondo spin-flip for various spin projections becomes easier [see formula (1.1)] and the suppression of the magnetic moment due to Kondo fluctuations wins out over the interaction between magnetic centers: $T_K \gg T_{\text{RKKY}}$.

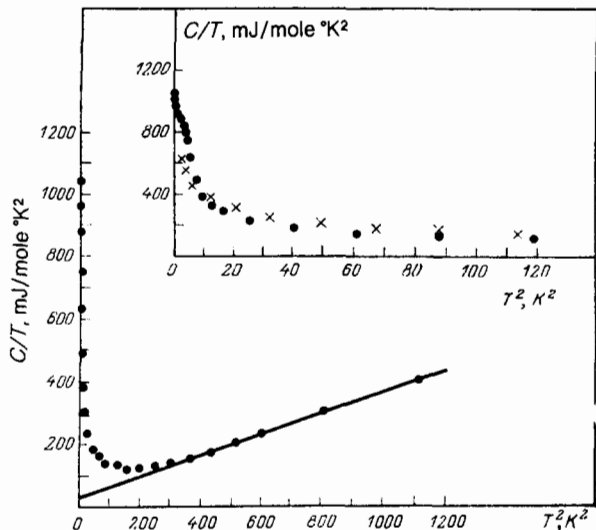


FIG. 9. Temperature dependences of the specific heat $C(T)$ of single-crystal CeCu_2Si_2 in the coordinates $C/T, T^2$.⁹⁵ The inset shows the low-temperature portion $C/T = f(T)$; x—data obtained in a field $H = 100$ kOe.⁹⁵

In order to understand the nature of the unusual low-temperature properties of NKs, one must appreciate the extreme importance of the fact that these systems must be considered as systems whose tendency to order magnetically is suppressed by the effect of Kondo compensation of the magnetic ions, i.e., $T_K \gg T_{\text{RKKY}}$. Moreover, the compounds CeCu_2Si_2 , CeAl_3 , CeCu_6 , as opposed to classical Kondo-impurity systems— $\text{Cu}(\text{Fe})$, $\text{Au}(\text{Fe})$, $\text{Cu}(\text{Mn})$, etc.—are *concentrated* Kondo systems with magnetic Ce^{3+} ions in every unit cell; therefore, in such CKS one can expect the appearance of ASRs of gigantic amplitude.

3. GIANT ABRIKOSOV-SUHL RESONANCES IN NONMAGNETIC KONDO LATTICES

3.1. Low-temperature anomalies in the electronic heat capacity of a nonmagnetic Kondo lattice

The temperature dependence of the specific heat $C(T)$ of a normal metal consists of a sum of electron (βT)³ and phonon (γT) contributions:

$$C(T) = \gamma T + \beta T^3. \quad (3.1)$$

The dependence of C on T , plotted against the coordinate $(C(T)/T) \cdot T^2$, allows us to determine the electronic coefficient γ as the value of C/T as $T^2 \rightarrow 0$, and the phonon coefficient β as the slope of the linear part of the function $C/T = f(T^2)$. The coefficient γ is proportional to the density of states at the Fermi level:

$$\gamma = \frac{\pi^2}{3} k_B^2 g(E_F). \quad (3.2)$$

For normal and transition metals, the value of γ is 1–10 mJ/mole K^2 in order of magnitude.⁹⁴ For the NKs CeCu_2Si_2 ⁹⁵ (Fig. 9), CeAl_3 ,³ CeCu_6 ,^{89,96} UBe_{13} ,^{3,7} the function $C(T)/T = f(T^2)$ for low temperatures slopes up⁴ towards the region of large values of C/T , and for $T \rightarrow 0$ the coefficient γ attains the gigantic value of 10^3 mJ/mole K^2 (Table II), almost a three orders of magnitude increase in the value of γ over that of a normal metal. We note that when we compare the coefficient γ in NKs containing several atoms per mole with the coefficient γ for simple metals with one atom per mole, it is necessary to divide the data presented in Table II for NKs by 5 (CeCu_2Si_2), 14 (UBe_{13}), etc.²¹ However, even after such a recalculation, the low-temperature electronic contribution to the heat capacity in NKs turns out to be 2–3 orders of magnitude larger than for normal metals. In NKs at low temperatures, an enhancement of the Pauli paramagnetism is also observed:

$$\chi_P(T) \approx \text{const} \approx \mu_B^2 g(E_F), \quad (3.3)$$

to first approximation (see Table II) the value of χ_P increases in NKs relative to a normal metal by the same factor as the growth in the coefficient γ , i.e., for the NKs CeCu_2Si_2 , CeCu_6 , CeAl_3 , UBe_{13} both quantities— χ_P and γ —vary essentially because of the increase in the density of states $g(E_F)$. Qualitatively,⁹⁷ the contribution of the renormalized density of states to the experimentally-observed value of χ_P can be characterized by the ratio χ_P/χ_P^0 where χ_P^0 is the value of the susceptibility evaluated using formulae (3.3) and (3.2):

$$\chi_P = 3\mu_B^2 \gamma \frac{\pi^2}{k_B}. \quad (3.4)$$

The ratio χ_P/χ_P^0 (see Table II) is of order unity, and consequently the low-temperature enhancement in χ and γ for NKs is connected primarily with a renormalization of the density of states $g(E_F)$, i.e., for NKs at low temperatures

TABLE II. A comparison of the characteristics of nonmagnetic Kondo lattices (CeCu_2Si_2 , CeAl_3 , CeCu_6 and UBe_{13}) and a normal metal (Cu).

Metal	$\gamma(T \rightarrow 0)$ mJ/mole K^2	$\chi(0)$ CGS/mole	$\sim A$ $\mu\Omega\text{-cm/K}$	$\frac{m^*}{m_0}$	T_F^* K	k_F 10^8 cm^{-1}	v_F cm/s	$\frac{R_H(4,2 \text{ K})}{R_H(100 \text{ K})}$
CeCu_2Si_2	1050 ⁹⁵	0.0065 ¹⁵⁵	10 ^{34,118}	500	8	0.74	$1.68 \cdot 10^5$	15 ^{54,58}
CeAl_3	1620 ⁶	0.036 ⁶	35 ⁶	800	5	0.73	$1.00 \cdot 10^5$	18 ^{59,60}
CeCu_6	1450 ⁸⁹	0.027 ⁹⁶	26.3 (i a) ⁹³ 13.8 (i b) ⁹³ 39.3 (i c) ⁹³	10^2-10^3	3	1.00	10^5	10 ⁶⁴⁻⁶⁶
UBe_{13}	1100 ⁷	0.015 ⁷		10^2-10^3	10	0.87 ¹⁷⁸	$3.4 \cdot 10^5$ ¹⁷⁸	12 ⁸⁰
Cu ⁹⁴	0.695	$10^{-8}-10^{-6}$	10^{-1}	0.1–1.0	$8 \cdot 10^4$	1.36	$0.57 \cdot 10^8$	1

there is in the vicinity of the Fermi level a narrow peak with a gigantic value of $g(E_F)$.

Since in the compounds CeCu_2Si_2 , CeAl_3 , CeCu_6 , UBe_{13} there is an electronic f shell, possessing an extremely small localization radius, it was initially assumed (see below, Fig. 15a) that the peak in the density of states $g(E_F)$ was connected simply with the exact coincidence of the narrow f band with the Fermi level, i.e., HFSs turn out to be intermediate-valence compounds. From this point of view, in HFSs based on cerium a significantly nonintegral cerium valence should be observed, $\nu(\text{Ce}) \approx 3.5$. However, the available experimental data (see the following section) show that the valence of cerium in CeAl_3 and CeCu_2Si_2 is close to integral: $\nu(\text{Ce}) \approx 3.0$, and the $4f$ band itself lies 1–2 eV below E_F and cannot be responsible for the giant value of the density of states at the Fermi level.

3.2. Valence state of f -ions in nonmagnetic Kondo lattices

Recently, the method of resonance photoemission⁹⁸ has often been used to determine the density of f states in the conduction band. In this technique, synchrotron radiation is used as a source of photons with energies rather close to the excitation of the deep and narrow $4d$ levels. In systems based on cerium, this energy is roughly equal to $\hbar\omega \sim 109$ eV. In varying $\hbar\omega$ near this value, a giant resonance in the $4f$ radiation⁹⁸ of cerium is observed at $\hbar\omega = 122$ eV. In the process of resonance photoemission a $4d$ electron is initially excited to the narrow $4f$ band, which has a large density of states:

$$4d^{10}4f^n(5d6s)^3 + \hbar\omega \rightarrow 4d^94f^{n+1}(5d6s)^3, \quad (3.5)$$

this intermediate excited state then relaxes, emitting an electron and screening the $4d$ hole in an Auger-type process:

$$4d^94f^{n+1}(5d6s)^3 \begin{cases} \rightarrow 4d^{10}4f^{n-1}(5d6s)^3 + e, & (3.6) \\ \rightarrow 4d^{10}4f^n(5d6s)^2 + e. & (3.7) \end{cases}$$

Relaxation via channel (3.6) leads to enhancement of the $4f$ emission, while decay via channel (3.7) leads to enhancement of radiation from the sd -valence band. Since the $4f$ band which participates in the intermediate process is extremely narrow, the resonant enhancements of the photoemission can be resolved separately, both for the sd and sd -valence states. Therefore, the resonant photoemission spectra can be used to estimate the width Δ_{4f} and position E_{4f} of the $4f$ band. According to the data⁹⁹ on CeCu_2Si_2 , $E_F - E_{4f} \sim 2.5$ eV and $\Delta_{4f} \sim 2$ eV, while for the compound CeAl_3 ,¹⁰⁰ $\Delta_{4f} \sim 2$ eV and $E_F - E_{4f} \sim 2$ eV.

Information about the magnitude of the Hubbard parameter U in NKLs can be obtained in isochromatic spectroscopy experiments using bremsstrahlung. In these experiments the energy interval is determined between the $4f^{n+1}$ and $4f^n$ states, which for CeAl_3 amounts to approximately 6–7 eV.¹⁰⁰

It must be noted that for the NKLs CeCu_2Si_2 ,⁹⁹ CeAl_3 ,¹⁰⁰ and UBe_{13} ,¹⁰¹ the resonance photoemission spectra also have a feature near E_F in addition to the peak at $E \approx E_{4f}$. The restriction of spectral resolution to the value 0.13–0.15 eV, does not permit a detailed investigation of the

feature at $E = E_F$. One possible explanation for a peak in the resonance photoemission spectrum at the Fermi level is the formation of a many-particle resonance in NKLs.^{101,102} As was shown in Refs. 100 and 103, the resonance photoemission spectra are exceedingly sensitive to the presence of adsorbed oxygen on the surface. Therefore, a correct measurement of the position of the $4f$ band can be carried out by cleaving samples in vacuum and at low temperatures, so that the diffusion of adsorbed oxygen out of the whole volume to the illuminated surface is negligibly small.¹⁰¹ For high temperatures, even a few minutes after cleaving so much diffusing oxygen has accumulated at the sample surface that the resonance photoemission spectra are noticeably distorted.

Unfortunately, even a correctly determined photoemission spectrum still does not give the energy dependence of the density of states, since the deep $4d$ holes (3.5) interact significantly with the charged $4f$ states $4f^1$ and $4f^2$; this leads to a marked downward shift in the $4f^1$ and $4f^2$ levels on the energy scale. In Ref. 104, a method was suggested for finding the $4f$ -band parameters of the model from the x-ray photoemission spectrum. Using Δ_{4f} and E_{4f} as fitting parameters, it was found that typical values of these parameters, even in compounds which are traditionally classified^{43,44} as IVC— CeRu_2 , CeNi_2 , CePd_3 —amount to $\Delta_{4f} \sim 0.1$ – 0.2 eV, $E_F - E_{4f} \sim 1$ – 2 eV.

The valence state of cerium in CeCu_2Si_2 , CeAl_3 and other cerium systems can also be found from data on the L_{III} x-ray absorption edge, by comparing the L_{III} absorption spectrum of the material under study with the spectrum of a “bench mark” compound with a well-known cerium valence. As a rule, cerium compounds with ionic bonds are chosen as bench marks, e.g., CeF_3 where $\nu(\text{Ce}) = 3 +$. For CeCu_2Si_2 ⁴⁵ and CeAl_3 ⁴⁶ this method gives a value of $\nu = 3$ – 3.05 .

To summarize, analysis of the available spectroscopic data on the NKLs CeCu_2Si_2 and CeAl_3 , and also of the “typical cerium IVCs” CePd_3 , CeNi_2 , CeRu_2 , etc., allows us to conclude that both in NKLs and in cerium IV compounds that valence state of cerium ions is realized which is close to the integral Ce^{3+} state, and not to the essentially nonintegral value $\nu(\text{Ce}) = 3.5$.

3.3. Investigation of dynamic spin fluctuations in nonmagnetic Kondo lattices based on experiments using quasielastic scattering of thermal neutrons

In contrast to methods such as x-ray photoemission, isochromatic spectroscopy, etc., which are sensitive to charge fluctuations, the method of quasielastic scattering of thermal neutrons with energies 3–180 meV allows us to investigate the dynamics of spin fluctuations in metals.¹⁰⁵ The usual quasielastic line in neutron scattering is observed against a background of high-amplitude peaks corresponding to elastic scattering by nuclei. These lines are approximated by Lorentzians with centers $\hbar\omega \neq 0$ and half-widths $\Gamma/2$, determined by the spin fluctuation time τ_{sf} : $\Gamma/2 \sim \tau_{sf}^{-1}$. The dependence of $\Gamma/2$ on temperature for CeCu_2Si_2 ⁷⁰ and CeAl_3 ¹⁰⁶ is shown in Fig. 10. Both compounds have finite widths Γ as $T \rightarrow 0$: $\Gamma/2(T \rightarrow 0) = 0.85$ meV for CeCu_2Si_2 and

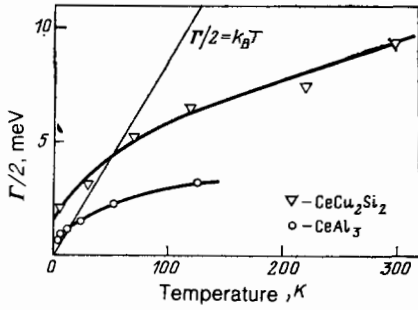


FIG. 10. Temperature dependences of the linewidth Γ of the quasielastic scattering of thermal neutrons for CeCu_2Si_2 ⁷⁰ and CeAl_3 .¹⁰⁶

$\Gamma/2(T \rightarrow 0) = 0.35$ meV for CeAl_3 . The dependence $\Gamma/2 = f(T)$ for these NKLs is analogous to the corresponding dependence for classical Kondo-impurity systems, i.e., $\text{Cu}(\text{Fe})$,^{107,108} and is described by the following empirical relation (see the continuous curve in Fig. 10):

$$\frac{\Gamma}{2} = \left(\frac{\Gamma}{2}\right)_{T=0} + aT^{1/2} \quad (3.8)$$

The fine line in Fig. 10 corresponds to the equation $\Gamma/2 = k_B T$ and divides the coordinate plane $\Gamma/2 - T$ into a high-temperature regime where the characteristic time τ_T for thermalization of spins is smaller than the time τ_{sf} for Kondo spin fluctuations: $k_B T > \Gamma/2$, $\tau_T < \tau_{sf}$, and a low-temperature regime $k_B T < \Gamma/2$ in which, conversely, the Kondo spin fluctuations are faster than the spin thermalization processes: $\tau_{sf} < \tau_T$. For high temperatures, when $\tau_T < \tau_{sf}$, the Kondo spin fluctuations from a state $S = 1/2$ to a state $S = -1/2$ occurring through a state at E_F are slowed in relation to τ_T . Therefore, a high temperature measurement is dominated by thermal fluctuations of uncompensated spin, and as a result the Curie-Weiss law is obeyed with an effective magnetic moment μ_{eff} close to the value corresponding to a free magnetic ion.

As the temperature falls, a crossover regime is reached from $k_B T > \Gamma/2$ to $k_B T < \Gamma/2$ and $\tau_{sf} < \tau_T$. In this case, the Kondo spin fluctuations are established at $T \rightarrow 0$ so rapidly compared to τ_T that within a time τ_T several spin flips occur for a $4f$ electron between the states $S = 1/2$ and $S = -1/2$. Consequently, both spin projections are equally probable, i.e., the rapid Kondo spin fluctuations lead to suppression of the effective magnetic moment: $\mu_{\text{eff}} \rightarrow 0$ as $T \rightarrow 0$.

The dynamic spin fluctuations described above in Kondo systems with $S = 1/2$ are retained also in the case $S > 1/2$, if in this case due to the crystal field splitting Δ_{CF} only the lowest doublet is active at low temperatures ($T_K \ll \Delta_{\text{CF}}$), which is analogous to a state with $S = +1/2$. This situation obtains in particular for the NKLs CeCu_2Si_2 ⁷⁰ and CeAl_3 ,⁷¹ where the sixfold degenerate level $j = 5/2$ of cerium is split into three doublets; the condition $T_K \ll \Delta_{\text{CF}}$ is fulfilled here. In Kondo systems with $j > 1/2$ and small values of the crystal field splitting, i.e., $\Delta_{\text{CF}} \ll T_K$, it is necessary to study spin fluctuations of Kondo type over all $2j + 1$ states. Then for low temperatures ($\tau_{sf} \ll \tau_T$) the occupation of all $2j + 1$ projections will be identical, which

leads to suppression of the effective magnetic moment as $T \rightarrow 0$.

The theory of quasielastic scattering of thermal neutrons in CKS, taking into account the splitting of the $4f$ level in the crystal field, was set forth in Refs. 92 and 93; for CKS in which the parameters T_K and Δ_{CF} are not too different, the authors predict a suppression of the crystal field splitting for low temperatures, where a nonmagnetic singlet state occurs leading to equal probability of spin "hopping" across all the projections $-j \leq m \leq j$. As a result of these Kondo spin fluctuations, as $T \rightarrow 0$ states with different quantum numbers m are "mixed," which in the opinion of the authors of Refs. 92 and 93 can cause an effective decrease in the crystal field splitting for $T < T_K$. This effect is apparently less important for CKS with strongly differing parameters T_K and Δ_{CF} .

3.4. Nonmagnetic Kondo lattices viewed as systems of N_i non-interacting Kondo impurities

Let us enumerate from the beginning the most characteristic properties of the typical NKLs CeCu_2Si_2 , CeAl_3 and CeCu_6 :

1. A gigantic low-temperature contribution to the electronic specific heat and enhancement of the low-temperature Pauli paramagnetism (see Table II).
2. Complete suppression of the effective magnetic moments of the Ce^{3+} ions as we go from $T \gg T_K$ to $T \ll T_K$: $\mu_{\text{eff}}^2 \sim \chi(T)$ as $T \rightarrow 0$.
3. The dominance of Kondo spin compensation over the tendency to undergo a magnetic transition due to the indirect RKKY interaction: $T_K \gg T_{\text{RKKY}}$.
4. A crossover regime—from slow spin fluctuations compared to τ_T at high temperatures to rapid spin fluctuations for low temperatures, i.e., suppression of the magnetic moment in NKLs related to fulfilling the condition $\tau_{sf} \ll \tau_T$ at sufficiently low temperatures.
5. Fermi-liquid behavior of the resistivity and thermoelectric-power coefficients: the coefficient $A(\Delta\rho = AT^2)$ and $a(S = -aT)$ are sharply enhanced as we go from the Kondo impurity to the Kondo lattice in the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$, $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$ and $\text{Ce}_x\text{La}_{1-x}\text{Cu}_6$.
6. Anomalous low-temperature growth in the Hall coefficient as T decreases from $T \gg T_K$ to $T \lesssim T_K$.

In all the properties we have enumerated, a characteristic parameter—the Kondo temperature T_K —plays a role. We can estimate this parameter by several independent techniques: from data on $\rho(T)$ (T_K^{ρ}); from neutron scattering ($T_K^{\Gamma} \sim \Gamma(0)/2$); from the dependence of χ on T (T_K^{χ}); from the negative magnetic resistance (NMR) minimum ($T_K^{\text{NMR}} = T_{\text{min}}/2$). For CeCu_2Si_2 , these estimates give

$$T_K^{\rho} \sim T_K^{\Gamma} \sim T_K^{\chi} \sim T_K^{\text{NMR}} \approx 8 \text{ K}, \quad (3.9)$$

which amount to 8 °K, 5 °K for CeAl_3 and 2° – 3° K for CeCu_6 .^{109,110}

The low-temperature NKL anomalies 1–6 point to the presence of a narrow ($\sim T_K$) peak in the density of states near E_F . This peak cannot be the $4f$ band itself (see below, Fig. 15b), since the valence of cerium in the NKLs CeCu_2Si_2

and CeAl_3 is almost integral, i.e., we cannot study NKLs as if they were IVCs with $\nu(\text{Ce}) \approx 3.5$. An analysis of the variation in the properties of the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$, $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$, $\text{Ce}_x\text{La}_{1-x}\text{Cu}_6$ shows clearly that the "genetic" anomalies of NKLs are connected with the Kondo processes, which distinctly appear for $x \ll 1$, and then smoothly lead to NKL anomalies for $x \rightarrow 1$.

For Kondo impurities, it was shown as early as 1965 by Abrikosov^{18,19} and Suhl²⁰ that it was possible for a narrow ($\sim T_K$) many-particle resonance to form in the scattering of electrons with energy $E \approx E_F$. In 1972, Grüner and Zawadowsky³⁶ suggested a semi-phenomenological model, within whose framework it was assumed that the scattering resonance leads to the appearance at the Fermi level of a narrow peak in the density of states, which they called an "Abrikosov-Suhl resonance." With the help of this model, they succeeded in describing the experimental data on the dependence of the Kondo contribution to the resistance for a number of transition elements in Kondo systems based on copper and aluminum.³⁶

In investigating NKLs as concentrated Kondo systems, it is natural to assume^{16,17,48,49} that the narrow peak in the density of states at the Fermi level of NKLs is an ASR, whose amplitude in the approximation of N_i non-interacting Kondo impurities must be enhanced compared to a single Kondo impurity by several times. Since in one mole of an NKL $N_i = N_A$, this enhancement can in principle completely account for the presence of the giant density of states at the Fermi level: $g^R(E_F) = N_i g_1^R(E_F) \gg g^0(E_F)$. For the Kondo Hamiltonian²⁶

$$H_{s-d} = \sum_{\mathbf{k}, \sigma=\uparrow, \downarrow} E_{\mathbf{k}} c_{\mathbf{k}\sigma}^{\dagger} c_{\mathbf{k}\sigma} + J \sum_{\mathbf{k}, \mathbf{k}', \sigma, \sigma'} \hat{S}_{\mathbf{k}\sigma}^{\dagger} \hat{S}_{\mathbf{k}'\sigma'} c_{\mathbf{k}\sigma}^{\dagger} c_{\mathbf{k}'\sigma'} \quad (3.10)$$

($c_{\mathbf{k}\sigma}^{\dagger}$, $c_{\mathbf{k}\sigma}$ are creation and annihilation operators for a state with wave vector \mathbf{k} and spin σ ; \hat{S} , \hat{s} are spin operators of the magnetic impurity and of a conduction-band electron), the exact Vignani-Andrej solution leads to the following expression for determining the low-temperature asymptotic specific heat and magnetic susceptibility:^{32,33}

$$\frac{c_v(T \rightarrow 0)}{T} = \gamma = \frac{\pi k_B}{6T_0} \quad (3.11)$$

$$\chi(0) = \frac{(\mu_B g)^2}{4\pi k_B T_0} \quad (3.12)$$

In formula (3.12), g is the Landé factor. The parameter T_0 is related to the Kondo temperature T_K by Wilson's constant \tilde{W} ¹¹¹:

$$\frac{T_K}{T_0} = \tilde{W} = 4\pi \cdot 0.1026 = 1.2902. \quad (3.13)$$

If the Kondo Hamiltonian (3.10) is generalized to impurities with spin $S > 1/2$, but, as before, we at the outset investigate only conduction-band electrons with zero orbital angular momentum l (as in the case $S = 1/2$), then under these circumstances the impurity with $S > 1/2$ is not fully compensated.¹¹²⁻¹¹⁴ Consequently, we cannot adequately describe the behavior of a magnetic impurity with $j > 1/2$ by a simple substitution $S \rightarrow j$ in (3.10), since it is well known⁴ that also for $j > 1/2$ the effective moment of a Kondo impurity $\mu_{\text{eff}}^2 \sim \chi T \rightarrow 0$ for $T \rightarrow 0$. The behavior of impurities with

$j > 1/2$ is better described by the Coqblin-Schrieffer model,¹¹⁵ in which spin flip of band electrons is taken into account; the latter are represented by a sum of partial waves with nonzero orbital angular momentum $l > 0$ over all spin projections $-j < m < j$:

$$H_{\text{CS}}^{\dagger} = \sum_{\mathbf{k}, m} E_{\mathbf{k}} c_{\mathbf{k}m}^{\dagger} c_{\mathbf{k}m} + J \sum_{\mathbf{k}, m, \mathbf{k}', m'} \sum_{m''} c_{\mathbf{k}'m'}^{\dagger} c_{\mathbf{k}m} a_{m''}^{\dagger} a_{m''}. \quad (3.14)$$

In this model, the exchange interaction between the conduction band electrons ($l > 0$) and the LMM (localized magnetic moment) ($j > 1/2$) leads to complete compensation of the effective impurity moment as $T \rightarrow 0$. The magnetic susceptibility and electronic specific heat for (3.14) were found within the framework of the Bethe ansatz:

$$\chi(0) = \frac{\nu(\nu^2 - 1)(\mu_B g)^2}{24\pi k_B T_0} \quad (3.15)$$

$$\frac{c_v(T \rightarrow 0)}{T} = \gamma = \frac{(\nu - 1)\pi k_B}{6T_0} \quad (3.16)$$

where $\nu = 2j + 1$. The ratio χ/γ depends on the constants μ_B , g , k_B , and ν :

$$\frac{\chi(0)}{\gamma} = \frac{\nu(\nu + 1)\mu_B^2 g^2}{4\pi^2 k_B^2} = \frac{[j + (1/2)](j + 1)}{\pi^2 k_B^2} \mu_B^2 g^2. \quad (3.17)$$

By substituting $j = 1/2$ in (3.15) and (3.16), we obtain the magnetic susceptibility and the coefficient γ for the Kondo model (3.11)–(3.12).

Since $j = 5/2$ in the NKLs CeCu_2Si_2 , CeAl_3 , CeCu_6 and UBe_{13} , it would seem that in order to find γ and χ one could use formulae (3.15) and (3.16), setting $j = 1/2$ in them. However, it is actually necessary to take into account the fact that the level $j = 5/2$ is split by the crystal field. For the NKLs CeCu_2Si_2 ⁷⁰ and CeAl_3 ,⁷¹ the character of this splitting has been studied in neutron-scattering experiments. The sixfold degeneracy of the $j = 5/2$ level is split into three doublets; the condition $T_K \ll \Delta_{\text{CF}}$ is fulfilled, and in elastic Kondo processes at low temperatures only the lowest states act effectively, which makes these states analogous to states with $j = 1/2$. In such a situation the NKLs CeCu_2Si_2 and CeAl_3 can be treated as "j = 1/2" CKSs with $N_i = N_A$.^{2,16,117} In the compound UBe_{13} the cubic symmetry implies that the f level must split into a quartet Γ_8 and a Kramers doublet Γ_7 . However, at this time (as far as we know) no one has been able to establish experimentally the relative positions of the Γ_8 quartet and Γ_7 doublet for UBe_{13} .

As was shown above, the occupation of the ASR at $T = 0$, which determines its position relative to the Fermi level E_F , equals $1/(2j + 1)$.³¹ Therefore, for "j = 1/2" CKSs with $T_K \ll \Delta_{\text{CF}}$, $1/(2j + 1) = 1/2$, and the position of the ASR is precisely at the Fermi level: $E_R = E_F$. The giant low-temperature enhancement of the density of states for the NKLs CeAl_3 , CeCu_2Si_2 is connected with precisely this circumstance. When the opposite relation $T_K \gg \Delta_{\text{CF}}$ is fulfilled, it is the total angular momentum j which enters into the formation of the ASR and formulas (3.15)–(3.16) must be used. In cerium CKSs with high Kondo temperatures $T_K \gg \Delta_{\text{CF}}$ and $j = 5/2$, the occupation of the ASR equals $1/(2j + 1) = 1/6$; consequently, the ASR is formed somewhat higher up.

To estimate the molar specific heat and magnetic susceptibility of the NKLs CeCu_2Si_2 and CeAl_3 using a model of noninteracting Kondo centers, we must set $j = 1/2$ in formulae (3.15)–(3.16) and multiply the result so obtained by Avogadro's number N_A . For CeCu_2Si_2 , such an estimate gives $\gamma^{\text{theor}} \approx 700 \text{ mJ/mole} \cdot \text{K}^2$ and $\chi^{\text{theor}} \approx 0.03 \text{ CGS/mole}$ for $T_K = 8 \text{ K}$. In the NKL CeAl_3 , $T_K = 5 \text{ K}$ and $\gamma^{\text{theor}} \approx 1120 \text{ mJ/mole} \cdot \text{K}^2$.

The values so obtained agree to within an order of magnitude with experimental data (see Table II). If we treat T_K as a fitting parameter, then with the help of a model of noninteracting " $j = 1/2$ " Kondo centers, we can arrive at a good description of the experimental variation in the specific heat $C(T)$ over a rather wide temperature interval¹¹⁸ (Fig. 11). For high temperatures, all the levels split by the crystal field contribute to $C(T)$ (Fig. 11); as a result, anomalies in $C(T)$ of Schottky type are obtained.

For the compound UBe_{13} , the experimental temperature dependence of the specific heat $C(T)$ for low temperatures is well described by introducing a Lorentzian resonance of width 12 K lying precisely at the Fermi level.²¹ Overhauser and Appel suppose that this resonance is the doublet Γ_7 broadened by hybridization and falling precisely on the Fermi level: $E(\Gamma_7) = E_F$, while the quartet Γ_8 lies rather far below the Fermi level: $E(\Gamma_8) < E(\Gamma_7)$. We adopt the contrary view that for UBe_{13} the doublet Γ_7 lies below the quartet Γ_8 , and the relation $T_K \ll \Delta_{CF}$ holds, i.e., UBe_{13} is a " $j = 1/2$ " NKL with an ASR lying precisely at the Fermi level: $E_R = E_F$. From this point of view, the good agreement between calculations²¹ and experimental data on the specific heat, magnetic susceptibility and magnetization is connected, apparently, with the fact that in the range $T/T_K < 10$ the Vignani-Andrej solution can be very accurately represented by calculations using the resonance model;^{34,35} the width of this resonance is determined by the Kondo temperature, while its amplitude in the range $T/T_K < 10$ can be considered to be independent of temperature, i.e., the calculation in Ref. 21 once more illustrates the correctness of the conclusions of Refs. 34, 35.

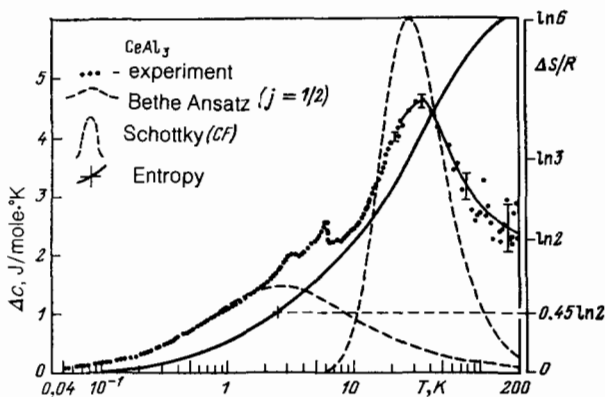


FIG. 11. Temperature dependence of the specific heat C for CeAl_3 .¹¹⁸ The dashed lines show calculations of the low-temperature contribution to the specific heat from the Kondo effect with " $j = 1/2$," and the high-temperature Schottky-type contribution from the f level split by the crystal field (CF).

Thus, determination of the character of the f level splitting for UBe_{13} in the crystal field serves as an experimental criterion allowing us to identify more precisely the nature of the peak in $g(E)$ at $E = E_F$ for this compound. If the Γ_7 doublet lies above the Γ_8 quartet, then apparently the Overhauser-Appel model is correct; if the opposite holds, i.e., $E(\Gamma_8) > E(\Gamma_7)$, then most likely UBe_{13} is a " $j = 1/2$ " NKL with $E_R = E_F$.^{2,16,117}

For dense Kondo systems, the degeneracy $\nu = 2j + 1$ of the f level is lifted by the crystal field and a three-level system is formed just as in CeAl_3 and CeCu_2Si_2 . The Kondo temperature is then determined by the following equation⁷²:

$$\left(\frac{T_K}{W}\right)^{\nu_0} \left(\frac{T_K + \Delta_{CF1}}{W + \Delta_{CF1}}\right)^{\nu_1} \left(\frac{T_K + \Delta_{CF2}}{W + \Delta_{CF2}}\right)^{\nu_2} = \exp\left[-\frac{1}{g(E_F)J}\right], \quad (3.18)$$

where the subscripts 0, 1, 2 refer to the lowest, first and second excited levels; ν_i is the degeneracy of the i th level; $\nu = \nu_0 + \nu_1 + \nu_2$ (for CeAl_3 , CeCu_2Si_2 , $\nu_0 = \nu_1 = \nu_2 = 2$), while Δ_{CF1} and Δ_{CF2} are the spacings between the lowest and the first excited levels, and between the lowest and the second excited levels, respectively; W is the half-width of the conduction band. Within the framework of the theory in Ref. 72, for high temperatures $T \gg \Delta_{CF1}$, Δ_{CF2} it is expedient to introduce a conduction-band Kondo temperature T_K^h :

$$T_K^h = W \exp\left[-\frac{1}{\nu g(E_F)J}\right]. \quad (3.19)$$

This expression also applies in the cases $T_K \gg \Delta_{CF1}$, Δ_{CF2} or $\Delta_{CF1} = \Delta_{CF2}$. As the temperature decreases to the region $T \ll \Delta_{CF1}$, Δ_{CF2} for $T_K \ll \Delta_{CF1}$, $\Delta_{CF2} \ll W$, we have

$$T_K = \left(\frac{W}{\Delta_{CF1}}\right)^{\nu_1/\nu_0} \left(\frac{W}{\Delta_{CF2}}\right)^{\nu_2/\nu_0} W \exp\left[-\frac{1}{\nu_0 g(E_F)J}\right]. \quad (3.20)$$

From formulae (3.19) and (3.20) we also obtain the formula $T_K^h = (T_K \Delta_{CF1} \Delta_{CF2})^{1/3}$, already given above for CeAl_3 , CeCu_2Si_2 and other NKLs with Δ_{CF1} , $\Delta_{CF2} \gg T_K$ and $\nu_0 = \nu_1 = \nu_2 = 2$. Thus, in " $j = 1/2$ " NKLs of the type CeAl_3 , CeCu_2Si_2 there are two Kondo temperatures— T_K and T_K^h , each of which is a characteristic energy parameter within its temperature interval.

Data on the Hall effect for NKLs^{54,59-61} can be used to estimate the Fermi velocity v_F , wave vector k_F and effective mass m^* (see Table II). For CeCu_2Si_2 , the Hall concentration for $T = 100 \text{ K}$ amounts to $n(100 \text{ K}) = 1.4 \times 10^{22} \text{ cm}^{-3}$, which is close to the corresponding value for LaCu_2Si_2 and to the value of $1.34 \times 10^{22} \text{ cm}^{-3}$ found from the condition that in CeCu_2Si_2 there is one electron per unit cell, that is the conduction band is half-full.

According to Landau's Fermi-liquid theory,¹⁰⁹ the strong interaction between electrons as $T \rightarrow 0$ does not change the magnitude of the wave vector k_F . Taking this circumstance into account, and assuming that the Fermi surface is spherical, we have

$$n = \frac{k_F^3}{3\pi^2}. \quad (3.21)$$

According to formula (3.18) for CeCu_2Si_2 , we find that $k_F = 0.745 \times 10^8 \text{ cm}^{-1}$ and, consequently,

$$m_h^* = \frac{3\gamma\hbar^2}{(k_F k_B)^2} = 512 m_0, \quad (3.22)$$

$$v_F = \frac{\hbar k_F}{m_h^*} = 1.68 \cdot 10^5 \text{ cm/s}, \quad (3.23)$$

$$T_F^* = \frac{m_h^* v_F^2}{2k_B} = 47 \text{ K}. \quad (3.24)$$

The estimates so obtained differ in certain ways from the analogous estimates in Ref. 120. However, it must be kept in mind that both sets of values for k_F , m^* , v_F , T_F^* are correct only to an order of magnitude, since they were derived using many simplifying assumptions. It is interesting to note that the parameters m_h^* , v_F , T_F^* in NKLs differ by 2 orders of magnitude from the corresponding parameters for normal metals; the Fermi velocity, for example, in CeCu_2Si_2 turns out to be even smaller than the sound velocity $v_s \approx 2 \times 10^5$ cm/sec. To interpret the Hall anomalies for NKLs,^{49,54,59-61,80} it is natural to suppose that the contribution from heavy fermions with $m_h^* \sim (10^2-10^3)m_0$ to the Hall voltage in fields $H < 50$ kOe is negligibly small, if in addition to the heavy fermions there are also mobile electrons with effective masses $m_l^* < m_0 < m_h^*$. Assuming that the total concentration of light and heavy electrons does not depend on temperature, i.e., $n(m_h^*) + n(m_l^*) = \text{const}$, the low-temperature growth in the Hall coefficient $R_H(T)$ (see Fig. 5) can be related to a reduction in the concentration of mobile electrons as the temperature decreases from $T \gg T_K$ to $T \ll T_K$. According to this interpretation, the low-temperature anomalous growth of $R_H(T)$ is a consequence of the increase in the effective mass of a significant fraction of the Fermi electrons due to the formation of the ASR with $E_R = E_F$.

An alternative interpretation of the anomalies in $R_H(T)$ in NKLs is based on the presence of enhanced nonsymmetric scattering⁷⁷ ("skew scattering") in NKLs. The Kondo spin compensation in CKSs leads to an increase in the density of electrons with nonzero orbital angular momentum $l > 0$ in a "Kondo cloud" around the magnetic centers, and likewise to a growth in the correction to the Hall coefficient because of nonsymmetric scattering.¹²¹ The theory constructed in Ref. 121 gives a rather good description of the available experimental data,^{48,54,59-61,64,65,78-80} including the change in sign of R_H in CePd_3 ,^{78,79} the growth in $|R_H|$ for NKLs^{48,54,59-61,64,65,80} and the sign $R_H(4.2 \text{ K}) > 0$ of the Hall coefficients in CeCu_2Si_2 , UBe_{13} , CeAl_3 , and CeCu_6 . The growth of the low-temperature values of $R_H(T)$ for the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ and $\text{Ce}_x\text{La}_{1-x}\text{Al}_3$ for $x \rightarrow 1$ (see Fig. 5), from this point of view, reflects the increase of the ASR as we approach the NKLs CeCu_2Si_2 and CeAl_3 .

Thus, the basic difference between the Kondo-impurity and Kondo-lattice systems is contained in the magnitude of the ASR: whereas for the Kondo impurity systems the ASR amplitude is small compared to the unperturbed density of states, in NKLs the ASR acquires a gigantic amplitude.

The width of the ASR is determined by the Kondo temperature $T_K = 1^{\circ}-10^{\circ}\text{K}$ [see also (3.18)–(3.20)], which for NKLs at low temperature is several orders of magnitude smaller than the electron-gas degeneracy temperature of

normal metals. Therefore, in NKLs the effective mass of the heavy fermions must attain values of $(10^2-10^3)m_0$.

The position of the ASR relative to E_F depends on the magnitude Δ_{CF} of the crystal-field splitting and on T_K . For "j = 1/2" CKSs with $\Delta_{CF} \gg T_K$, the ASR for $T < T_K \ll T_K^h$ forms exactly at the Fermi level. Thus, we expect that to create a giant density of states at the Fermi level it is also necessary to have a CKS in which the lowest state split off by the crystal field is a doublet, and in which the condition $\Delta_{CF} \gg T_K \gg T_{RKKY}$. That is, $N_i \sim N_A$, i.e., the concentration of Kondo centers is large; however, the indirect magnetic exchange interaction between magnetic ions must be suppressed while the resonance itself lies at $E_K = E_F$.

In fulfilling the condition $T_K \gg T_{RKKY}$ in CKSs, a fundamental role is played by two requirements—the exchange interaction J (under the assumption that $\nu = 2j + 1 = \text{const}$) must be strong enough to guarantee that the exponential dependence (1.1), which determines T_K , can win out over the quadratic dependence of T_{RKKY} on J , and also the degree of degeneracy ν of the f level itself.^{72,122} The critical value J_c found from the condition $T_K(J_c) = T_{RKKY}(J_c)$ (see Figs. 1, 4), according to Ref. 122, must reduce to zero as $\nu \rightarrow \infty$, i.e., for a very large number of projections $-j < m < j$ the local Kondo spin fluctuations which lead to spin flips for all m win out over the magnetic inter-center RKKY interaction even for arbitrarily small values of J .

Comparing the low-temperature properties of "j = 1/2" NKLs (see Table II) with the properties of normal metals, we can see differences of 2–3 orders of magnitude for those quantities which are directly connected with the existence of a peak in $g(E)$ for $E = E_F$. This circumstance allows us to regard "j = 1/2" NKLs as a new class of metallic systems, in which low-temperature Kondo-type interelectron correlations, which correspond to a certain effective attraction between electrons, lead to the formation near E_F of a narrow ASR of gigantic amplitude. To first approximation, NKLs for $T \lesssim T_K$ can be studied as ensembles of N_i noninteracting Kondo singlets. Estimates of $\chi(0)$, $\gamma(T \rightarrow 0)$, T_F^* , m_h^* and the Hall coefficient $R_H(T)$ using such a model gives values which agree to an order of magnitude with the corresponding experimental data. For a more adequate description of the low-temperature properties of NKLs, it is necessary to include coherent Kondo spin fluctuations for $T < T_{coh} < T_K$.

3.5. Coherent effects in nonmagnetic Kondo lattices for $T \ll T_K$

For sufficiently low temperatures $T < T_{coh}$ we can apply Bloch's theorem to a periodic CKS, i.e., a Kondo lattice; that is, in a system of periodically-located Kondo centers, coherence effects among the Kondo spin fluctuations must arise. When these coherence effects are present, the heavy fermions form a band with a well-defined dispersion law $E(\mathbf{k})$, characterized by the presence of a very large value of the effective mass. To investigate the distinctive features of the transformation of the Fermi surface when a heavy-fermion band forms, Martin^{123,124} has suggested that we use the

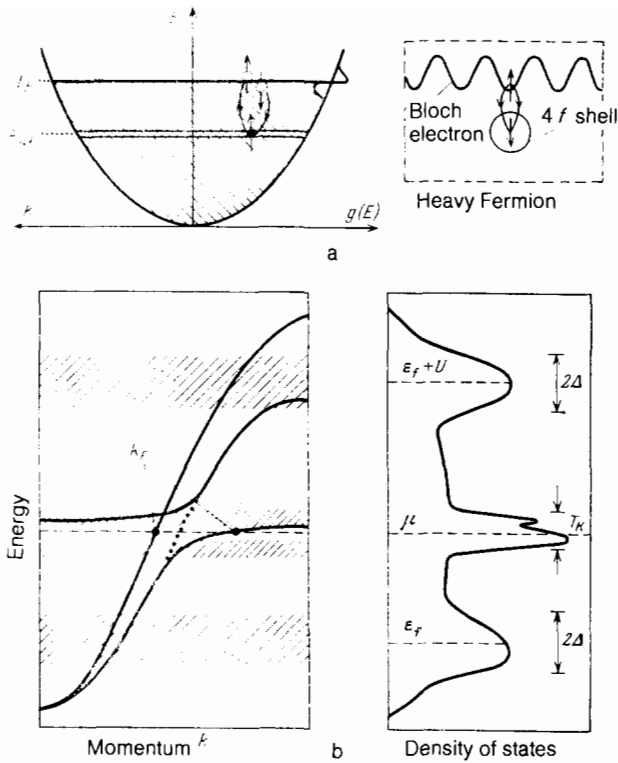


FIG. 12. (a) Low-temperature Kondo spin fluctuations leading to the formation of an ASR near E_F and to an increase in the effective mass of Fermi-level electrons.² (b) Behavior of the density of states (right) and energy spectrum (left) for a nonmagnetic Kondo lattice.¹²³ The scale T_K is magnified for clarity; the left side shows schematically the dispersion law for two directions in the Brillouin zone; along one of them, there are no massive Fermi-level electrons, while along the other the electrons are massive; all told, the behavior of the density of states in a nonmagnetic Kondo lattice is the same as for a system of Kondo impurities, except for a gap at E_F which is a consequence of the periodicity of the Kondo centers.

Luttinger theorem. A heavy fermion (Fig. 12) can be considered as a kind of "mixed" combination of localized f states and Fermi sd electrons. The formation of such a state on the one hand leads to effective suppression of the local magnetic moment of the f shells due to the rapid spin fluctuations; on the other hand it causes a sharp increase in the mass of the Fermi electrons themselves due to their significant "mixing" with the localized f electrons.

Significant anisotropy is predicted for a periodic system, i.e., an NKL (Fig. 12b): for some directions of the quasimomentum k there must occur a sharp increase in the effective mass of the Fermi electrons, while for other directions the change in the dispersion law is very small.¹²³⁻¹²⁵ As a result, in NKLs with $T < T_{\text{coh}}$ we expect that a local gap will appear at the Fermi level.¹²³⁻¹²⁸ If there is exactly one electron in the conduction band of a NKL for each magnetic center ($n_m = 1$), then a situation is possible where for $T < T_{\text{coh}} < T_K$ each electron forms a quasibound singlet state at "its" Kondo center, and then, apparently, the pseudogap at E_F must become a real gap. The authors of Refs. 126-128 are led to the conclusion that NKLs with $n_m = 1$ are insulators for any J ,^{126,127} or only for $J > J_c$.¹²⁸ For $n_m \neq 1$, an isolating gap at the Fermi level does not form; however, a

local minimum for $E = E_F$ in the background ASR does remain (see Fig. 12b). The appearance of fine structure in the ASR is connected with the transition from the incoherent Kondo-singlet regime for $T_{\text{coh}} < T < T_K$ to the regime of coherent NKL Kondo fluctuations for $T < T_{\text{coh}}$. Apparently, we can compare the local gap in the ASR with the Coulomb gap¹²⁹ in an impurity band in doped semiconductors—in both cases a reduced density of states at the Fermi level is connected with electron-electron correlations of the effective-repulsion type.

Variation in the density of states at the Fermi level in NKLs as we go to the coherent regime was studied experimentally in Refs. 130, 131. In NKLs the transition to the coherent regime leads to a decrease in $C/T \sim \gamma \sim g(E_F)$ (Fig. 13). The disruption of coherence which accompanies replacement of cerium in the NKL CeCu_2Si_2 , CeAl_3 by yttrium and lanthanum^{130,131} causes a suppression of the sections of decreasing $C/T \sim \gamma \sim g(E_F)$ (Fig. 13). These experiments show that destroying the periodicity in the positions of the cerium atoms by dilution with yttrium or lanthanum^{130,131} induces a transition to an ASR without a local pseudogap at the Fermi level: $C/T \approx \text{const}$ for $T < T_{\text{coh}}$. Based on the data from Refs. 130, 131, we deduce a characteristic temperature T_{coh} of roughly $T_K/10$.

The appearance in NKLs of a pseudogap at $E = E_F$, in agreement with the data of Ref. 118, leads to a sign change in the thermoelectric-power coefficient within the region $T < T_{\text{coh}}$ for CeCu_2Si_2 , CeAl_3 . In this case also, the loss of periodicity of the Kondo centers caused by the substitution of La or Y for Ce leads to suppression of the thermoelectric-power anomalies around $T_K/10$.⁵⁾

For the NKL CeAl_3 the transition from the incoherent regime to the coherent regime is accompanied by a change in

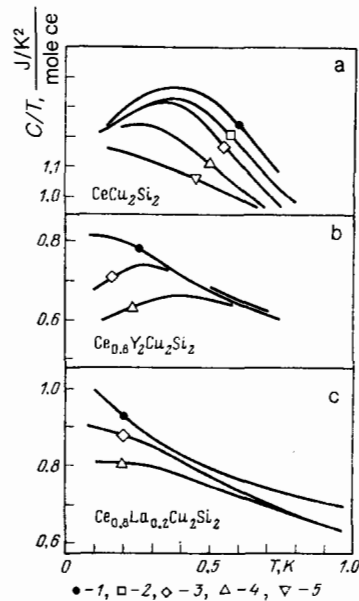


FIG. 13. Temperature dependences of the specific heat C for CeCu_2Si_2 (a), $\text{Ce}_{0.8}\text{Y}_{0.2}\text{Cu}_2\text{Si}_2$ (b), $\text{Ce}_{0.8}\text{La}_{0.2}\text{Cu}_2\text{Si}_2$ (c) in a magnetic field $H(\text{kOe}) = 0$ (1), 10 (2), 20 (3), 40 (4), and 80 (5).^{130,131}

sign of the thermal expansion coefficient.¹³² As a result of this, for $T < 1$ °K the electronic Grüneisen parameter is negative: $\Omega_G(\text{CeAl}_3) = -200$. By comparison, in the normal phase of ^3He , $\Omega_G = -1.1$ ($p = 0$) and $\Omega_G = -2.5$ ($p = 32$ bars).

Thus the Fermi-liquid behavior of the electronic system in the normal NKL CeAl_3 is qualitatively analogous to the behavior of liquid ^3He in its normal phase.

3.6. Compounds with variable valence as concentrated Kondo systems with high Kondo temperatures

In the NKL CeCu_2Si_2 , under the action of pressure there occurs a transformation of the temperature dependences of the resistivity from the curves $\rho(T)$ with Kondo growth in ρ , a maximum at $T = T_{\text{max}}^{(1)} \approx 10$ °K and Fermi-liquid decrease in ρ according to $\Delta\rho = AT^2$ for $T < T_{\text{max}}^{(1)}$ (Fig. 14) to dependences of $\rho(T)$ ($p > 40$ kbars) with metallic behavior of $\rho(T)$ over the entire range of temperature $T \lesssim 300$ °K.¹³³⁻¹³⁵ An analogous change in the dependence of $\rho(T)$ under pressure is also seen in CeAl_2 ,¹³⁶ CeIn_3 ,¹³⁷ and as a function of composition x in $\text{Ce}(\text{Rh}_x\text{Pt}_{1-x})_3$,^{138,139} $\text{Ce}(\text{In}_{1-x}\text{Sn}_x)_3$,¹⁴⁰ $\text{Ce}(\text{Sn}_x\text{Pb}_{1-x})_3$,¹⁴¹ $\text{Ce}(\text{Rh}_{1-x}\text{Ru}_x)_2$,¹⁴² and $\text{Ce}(\text{Ni}_x\text{Cu}_{1-x})_3$.¹⁴³ Usually such a transition in a CKS is treated as a transition CKS—to—IVC—to—a metal with cerium in a quadrivalent state. It is assumed that external pressure or controlled variation in composition significantly changes the position of the f level itself relative to E_F (Fig. 15a), from integer valence of the RE (the CKS regime) through a nonintegral RE valence (the IV regime) to a normal metal with an empty $4f$ shell. However, serious difficulties have been encountered recently when this model (the "promotion model") is used to interpret a whole range of experimental data. Let us assume that the IV anomalies which appear in "weak perturbation" experiments (i.e., measurements of magnetic susceptibility, specific heat, thermoelectric power, resistivity, Hall coefficient, etc.) are connected with coincidence of the narrow $4f$ band with the Fermi level, and let us use the width Δ_{4f} and position ($E_F - E_{4f}$) of this band as fitting parameters. Then a satisfactory description of the results of the "weak perturbation" measurements is obtained only for values $\Delta_{4f} < 0.01$

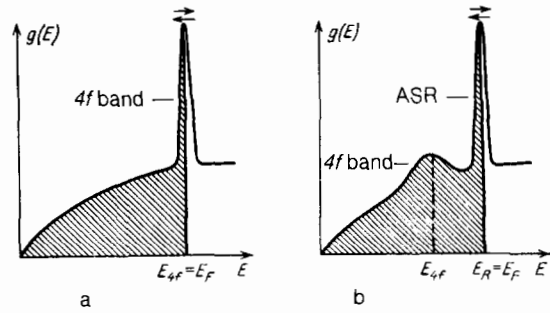


FIG. 15. A schematic illustration of the behavior of the density of states in a concentrated Kondo system.¹⁶ (a)—the "promotion model"; the peak in $g(E_F)$ is explained by the coincidence of the $4f$ band with the Fermi level. (b)—the new model in whose framework the peak $g(E_F)$ is an Abrikosov-Suhl resonance; the $4f$ band itself lies appreciably deeper below E_F , and has greater width.

eV and $E_F \approx E_{4f}$. In addition, for various cerium IVCs the valence states of cerium $v(\text{Ce})$ must include the entire range from 3 to 4 in this case. However, the values of Δ_{4f} , $E_F - E_{4f}$ and $v(\text{Ce})$ estimated in this way are found to be in contradiction with the data obtained with the help of spectroscopic methods, in which the system under study is rather strongly perturbed. In these "strong perturbation" experiments, the cerium valence $v(\text{Ce})$ in metallic CKSs never exceeds the value $v(\text{Ce}) = 3.25$ to 3.29 ,^{46,144,145} while a considerably larger width Δ_{4f} of (0.1–1.0) eV is obtained along with a submergence depth $E_F - E_{4f} \sim 1 - 2$ eV.^{98-104,146,147}

What are the actual parameters of the $4f$ band? Does the feature in the density of states coincide with the Fermi level and have a width of 0.1 eV, or is this feature located far below E_F and broadened to $\Delta_{4f} \sim 0.1 - 1.0$ eV? As the detailed analysis given in Ref. 2 shows, both groups of experiments mentioned above reflect the actual state of affairs: in both "weak perturbation" measurements and spectroscopic experiments it is clear that there are *two different* features in the density of states: a narrow band near E_F , which is the ASR, and a band with width 0.1–1.0 eV lying 1–2 eV below E_F , which is the $4f$ band itself (Fig. 15b). This interpretation removes the contradiction between the spectroscopic data and data obtained in "weak perturbation" experiments.

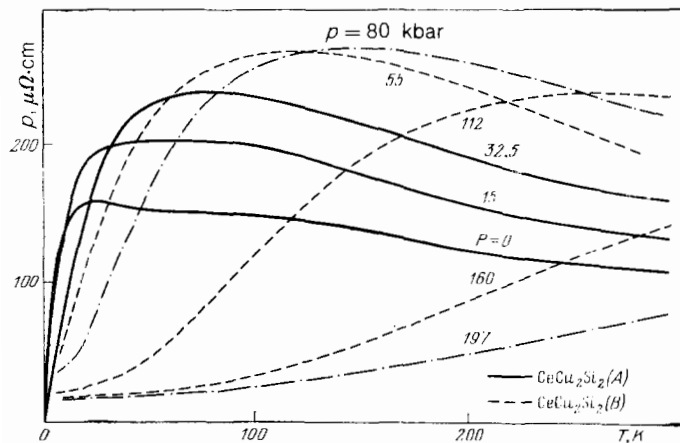


FIG. 14. Temperature dependences of the resistivity $\rho(T)$ for two samples of CeCu_2Si_2 (A, B) under pressure.¹³⁵ The low-temperature regions of all the curves of $\rho(T)$ are well linearized in the coordinates ρ, T^2 .

The character of the temperature variation $\rho(T)$ (see Fig. 14) in CeCu_2Si_2 under pressure, which usually is treated within the framework of traditional models as a transition from NKLs to IVCs, actually admits an alternative interpretation: the pressure-induced transformation of the curves $\rho(T)$ is a consequence of a shift in the maximum of $\rho(T)$ to the right on the temperature scale (see Fig. 14). In this case the Kondo temperature increases from $8^\circ\text{K} \ll \Delta_{\text{CF}}$ at $p = 0$ to $T_K \sim 300^\circ\text{K} \gg \Delta_{\text{CF}}$ for $p > 100$ kbar. In such a situation the metallic behavior of $\rho(T)$ for $p > 100$ kbar can be regarded as the region $\Delta\rho = AT^2$ observed below the maximum in $\rho(T)$, which for high pressures shifts into the region $T > 300^\circ\text{K}$ and is no longer visible for $T < 300^\circ\text{K}$. The transition induced by variation of the external parameters from a “ $j = 1/2$ ” CKS with $T_K \ll \Delta_{\text{CF}}$ and $E_R = E_F$ to a “ $j = 5/2$ ” CKS with $T_K \gg \Delta_{\text{CF}}$ and $E_R \neq E_F$ leads to a shift in the ASR relative to E_F which also imitates the CKS-IVC transition in which the $4f$ band itself actually is shifted very insignificantly while the ASR experiences a significant shift (see Fig. 15).

One of the weightiest arguments in support of viewing IVCs, or at least cerium-based IVCs, as CKSs with high Kondo temperatures $T_K > \Delta_{\text{CF}}$ is the existence of a limiting valence for cerium in metallic systems: $v(\text{Ce}) \leq 3.25 - 3.29^{46,144,145}$ (according to the data of Ref. 148, $v(\text{Ce}) \leq 3.40$). Since in order to describe data obtained from “weak perturbation” experiments used to study cerium IVCs, the narrow peak in the density of states must be placed somewhat above E_F ,² this would give a value of cerium valence in these compounds of 3.5–4.0 if the peak $g(E_F)$ were connected with the $4f$ level itself. It is obvious that the latter assumption contradicts the data on $v(\text{Ce})$.^{46,144,145,148}

Does all this imply that there are no IVCs in general? Of course, one cannot answer this question affirmatively, since it is well-known^{23,37,149,150} that when a certain predetermined relation between Δ_{4f} and U is fulfilled (in the case of nonintegral valence), the ASR and $4f$ band merge into a single peak near E_F , i.e., for small concentrations of noninteracting $4f$ impurities a regime of intermediate valence clearly exists. However, in going over to concentrated systems a state with essentially nonintegral valence can turn out to be thermodynamically unstable relative to a transition of the rare earth element to an almost integral valence state. Such a transition can be a consequence of the tendency to establish such a valence of the RE, which would ensure the maximum gain in free energy due to Kondo condensation processes. If this transition is discontinuous, it is called a “Kondo collapse.”^{50,51} It is interesting that, for example, in metallic cerium the “ejection” of the $4f$ band above the Fermi level is not possible even in the case of strong hydrostatic compression at $p < 200$ kbar, because it is more advantageous (due to lattice reconstruction) to retain the $4f$ level some distance below E_F , so as to have the optimal gain in free energy for each reconstruction due to Kondo condensation.

Thus, in a number of cases it is more appropriate to describe cerium “IVCs” as “ $j = 5/2$ ” CKSs with a $T_K > \Delta_{\text{CF}}$ and an ASR which is $1/6$ occupied and thus lies

above E_F . For these compounds, the traditional terminology “IVC” does not reflect adequately the essence of the processes which determine the nature of the peak near E_F . Therefore it seems expedient to introduce a new term—“ $j = 5/2$ ” CKSs, or high- T_K CKS—which would underline the nature of the feature in $g(E_F)$ as an ASR arising as a consequence of Kondo spin fluctuations.

A more detailed investigation of IVCs as CKSs with a high T_K can be found in Ref. 2.

4. SUPERCONDUCTING KONDO LATTICES

4.1. Superconductivity in CeCu_2Si_2

One of the surprising properties of the NKL CeCu_2Si_2 is heavy-fermion superconductivity, discovered in 1979 by Steglich and coauthors.⁵ Using the phase diagram of magnetic properties obtained for the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ (Fig. 6), one can trace the genesis of the unusual properties of CeCu_2Si_2 as it goes from a normal metal ($x = 0$) to a NKL ($x = 1$). The crossover regime—from magnetic ($T_K \ll T_{\text{RKKY}}$) to nonmagnetic ($T_K \gg T_{\text{RKKY}}$) (see Figs. 1, 4)—leads to the situation that in these alloys for $x \rightarrow 1$ the Kondo spin compensation dominates over the tendency to magnetic ordering via the indirect RKKY interaction. Therefore, the temperature of the magnetic transition (Fig. 16) falls sharply as $n \rightarrow 1$ and polycrystals of CeCu_2Si_2 with giant ASRs go into the superconducting state. A superconducting transition is accompanied by a jump in the specific heat, the appearance of anomalous diamagnetism and the reduction of the resistance to zero at $T = T_c$.⁵ It should be noted that the jump in specific heat at $T = T_c$ is of the same order of magnitude as the specific heat of CeCu_2Si_2 up to the transition, i.e., it is specifically the heavy fermions that are responsible for superconductivity in CeCu_2Si_2 .

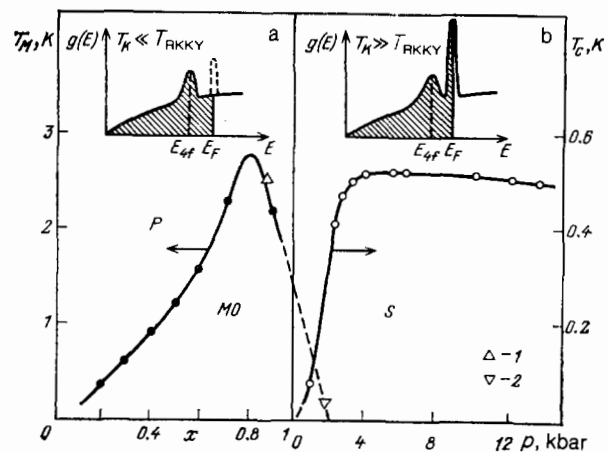


FIG. 16. Phase diagram of the magnetic and superconducting properties of the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ for $0 < x < 1$ and CeCu_2Si_2 at pressures up to 13.4 kbars.^{17,54} P denotes the paramagnetic phase, S the superconducting phase and MO the magnetically-ordered phase (i.e., a state with frozen-in spins). The inset illustrates the behavior of the density of states in two limiting cases: (a) the magnetic case $T_K \ll T_{\text{RKKY}}$ and (b) the nonmagnetic case $T_K \gg T_{\text{RKKY}}$. Starting from the assumption that varying the copper content in CeCu_2Si_2 is analogous to varying the pressure, we also present the data from Ref. 157 (1—a sample with a copper deficiency) and Ref. 155 (2—a sample with a copper excess).

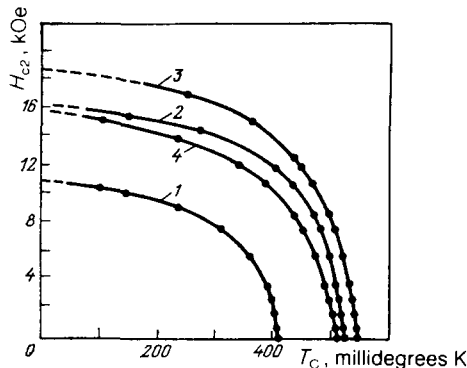


FIG. 17. Curves of the critical fields $H_{c2}(T_c)$ for $\mathbf{H} \parallel ic$ (i —current, c —tetragonal axis) for monocrystalline CeCu_2Si_2 for various pressures (in kbars): 2.2 (1), 3.3 (2), 3–5.0 (3) and 12.0 (4).⁵⁶

Under hydrostatic compression of CeCu_2Si_2 , along with increasing x in the series $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$, there occurs an increase in the Kondo temperature T_K . Therefore, on a phase diagram of magnetic and superconducting properties (Fig. 16), the variation of T_M for the alloys $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$ and T_c for the boundary compound CeCu_2Si_2 under pressure can be studied on a single $x-p$ scale, along which a displacement from left to right increases the magnitudes of J and T_K . Knowing the rate of displacement of the low-temperature maximum in $\rho(T)$ at $T = T_{\text{max}}^{(1)}$ under pressure and for variation of composition, the scale along the pressure axis (Fig. 16) can be chosen so that it ensures equality of the derivatives $\partial T_{\text{max}}^{(1)}/\partial p$ and $\partial T_{\text{max}}^{(1)}/\partial x$.

On the phase diagram (Fig. 16), the NKL CeCu_2Si_2 lies in a narrow critical region corresponding to the transition from the magnetic state to superconductivity. This circumstance leads to a strong dependence of the properties of CeCu_2Si_2 on external parameters—pressure, changes in composition and even on the method of preparation and subsequent thermal treatment. Stoichiometric single crystals, as opposed to polycrystalline samples, do not enter the superconducting state at normal pressures, at least in the range of $T > 0.02^\circ\text{K}$,^{48,49,56,95,151–154} but become superconducting only when a pressure $p > 1$ kbar is applied [Fig. 16: depen-

dence $T_c(p)$]. At the same time, single crystals of CeCu_2Si_2 grown from a composition with excess copper become superconducting even at $p = 0$ for $T_c = 0.5–0.65^\circ\text{K}$.^{120,153,155–157}

Data obtained in Ref. 156 give additional information on the characteristic behavior of the functions $T_M(x)$ and $T_c(x,p)$ (Fig. 16) near $x = 1$; for single-crystal CeCu_2Si_2 with excess copper (i.e., with large values of J and T_K) one observes re-entrant behavior in a constant magnetic field as the temperature falls, pointing to the possibility of an intersection of the functions $T_M(x)$ and $T_c(x,p)$. This implies that the magnetism of the Kondo—reduced LMM (local magnetic moment) in NKLs can in principle exist along with heavy-fermion superconductivity.

An investigation of the curves for the critical field $H_{c2}(T_c)$ in CeCu_2Si_2 (Fig. 17) shows that the derivative $-dH_{c2}/dT(T = T_c)$ is anomalously large, while the ratio $H_{c2}(0)/T_c(0)$ for $T_c \approx 0.5^\circ\text{K}$ exceeds by more than a factor of two the paramagnetic limit (Table III).^{151,152,48,49,56,120,155,156}

In the limit $l \ll \xi_0$ (l is the mean free path and ξ_0 the coherence length), the quantity dH_{c2}/dT at $T = T_c$ is determined by the product of the electron specific heat coefficient and the residual resistance ρ_0 :

$$\left. \frac{dH_{c2}}{dT} \right|_{T=T_c} = 5.26 \cdot 10 \left(\frac{\text{erg}}{\text{cm}^3 \text{K}^2} \right) \rho_0 (\text{Ohm} \cdot \text{cm}). \quad (4.1)$$

An estimate using formula (4.1) gives the values 100–200 $\text{kOe}/^\circ\text{K}$, which is in good agreement with the experimental data (see Table III). The correlation between the giant value of γ and $-dH_{c2}/dT(T = T_c)$, just as the value of the jump ΔC in specific heat at $T = T_c$, shows that superconductivity in CeCu_2Si_2 is connected with the heavy-fermion system formed by the narrow ($\sim 10^\circ\text{K}$) ASR. The high critical field $H_{c2}(0)$, which is proportional to the inverse Fermi velocity $(v_F^*)^{-1}$, indicates that v_F^* in CeCu_2Si_2 is anomalously small (see Table III).

The typical superconducting characteristics of CeCu_2Si_2 are determined in Ref. 120 for samples with very low residual resistances $\rho_0 = 3.5 \mu\Omega\text{-cm}$: $\xi_0 = 1.9 \times 10^{-6}$ cm, the London penetration depth $\lambda = 2 \times 10^{-5}$ cm,

TABLE III. Characteristics of heavy-fermion superconducting systems compared with the “normal” superconductor Sn.

Superconductor	$\gamma(T=0)$ mJ/mole $^\circ\text{K}^2$	$T_c, ^\circ\text{K}$	$H_{c2}(0),$ kOe	$H_{c2}(0)/T_c(0)$ kOe/ $^\circ\text{K}$	$-dH_{c2}/dT$ ($T = T_c$) kOe/ $^\circ\text{K}$	$\xi_0, \text{\AA}$ (coherence length)	$\lambda, \text{\AA}$ (London penetration depth)	H_{c2}, Oe (lower critical field)
CeCu_2Si_2	1050 ⁹⁵ 1000 ⁵ 1100 ⁷	0.04–0.6 ¹⁶¹ 0.5 ⁶ 0.85 ⁷	10–18 ⁵⁴	20–40 ⁵⁶	100–250 ⁵⁶ 60 ¹²⁰ 257 ⁷ 350 ¹⁷⁴ 275 ¹⁷⁸	190 ¹²⁰ 142 ¹⁷⁸	2000 ¹²⁰ 142 ¹⁷³	23 ¹²⁰
UPt_3	450 ¹⁰ 422 ¹⁹⁷	0.54 ¹⁰ 0.52 ¹⁷⁷ 0.49 ¹⁹⁷	> 16 ¹⁷⁷	> 32 ¹⁷⁷	63 ¹⁷⁷ 44 ¹⁹⁷	120 ¹⁷⁷ 170 ¹⁷⁷ 200 ¹⁹⁷	3600 ¹⁹⁷	22 ¹⁹⁷
Sn ⁹⁴	1.78	3.733	0.3	0.08	0.14			

the mean free path $l = 1.2 \times 10^{-6}$ cm, the lower critical field $H_{c1} \approx 23.3$ Oe. The data presented must be considered as estimates, since they vary from sample to sample; in addition, simplifying assumptions were used to obtain them. For example, it was assumed¹²⁰ that for low temperatures the anisotropies in CeCu_2Si_2 decrease and become negligibly small. This assumption contradicts the available experimental data^{154,157} for single-crystal CeCu_2Si_2 , in which the low-temperature anisotropy of the resistance in the basal plane compared to the resistance perpendicular to it equals 3–4.

4.2. The effects of pressure and controlled compositional variation on superconducting and normal characteristics of CeCu_2Si_2

The effect of pressure on the properties of the NKL CeCu_2Si_2 was studied for the first time in Refs. 48, 49, 54, 56, 134, 151, 152, and later by Bellarbi and coauthors.¹³⁵

Since the properties of CeCu_2Si_2 vary significantly from sample to sample, there is interest in analyzing data obtained from measurement of the various characteristics on the same sample, using pressure as an external parameter (Fig. 18). Enhancements in the low-temperature anomalous Hall coefficient and thermoelectric power coefficient are observed when pressure is applied; the Hall coefficient R_H (4.2 °K) and thermoelectric power coefficient $S(T) = T_{\text{extr}}^{(1)}$ pass through maxima at roughly the same pressures at which the values of T_c and $H_{c2}(0)$ attain their maximum values. This correlation in the variation of normal and superconducting properties with pressure indicates that the ASR apparently has its maximum amplitude at a pressure of 4–5 kbars.

In order to interpret the data obtained for CeCu_2Si_2 under pressure, it is necessary to take into account the following facts:^{2,117}

1. As suggested by spectroscopic data,^{99,100,103,158} the variation in J and T_K for cerium CKSs occurs, apparently, in large part, because of changes in the hybridization of the $4f$ and sd states. For CeCu_2Si_2 this hybridization depends

essentially on the position of the $3d$ band of copper relative to the $4f$ level.⁹⁹

2. Occupying an intermediate position between magnetic and nonmagnetic CKSs, the compound CeCu_2Si_2 must be very sensitive to contributions to the free energy from Kondo condensation processes, which can lead to enhancement of the effectiveness of the phonon mechanism of attraction between heavy fermions.¹⁵⁹

3. Variation of T_K under pressure can lead to a shift in the characteristic temperature T_{coh} and, consequently, to a pressure dependence of the parameters which give rise to the local gap formed at the ASR for $E = E_F$ in the coherent regime $T < T_{\text{coh}}$.

4. Under compression, it is possible to decrease the magnitude of the $4f$ level splitting in the crystal field, due to the pressure-induced decrease in the anisotropy. This effect in turn affects the relation between T_K and Δ_{CF} , and influences the position of the ASR.

5. We also cannot exclude from the investigation the pressure dependence of the number of electrons n_m at a magnetic center, since the depth of the pseudogap at the ASR for $E = E_F$ depends on n_m .

Unfortunately, the available experimental data do not allow us to determine what factors are dominant. Nevertheless, the behavior of T_c under pressure can in general terms be interpreted in the following way¹⁵⁷: the growth of T_c in the region $p < 5$ kbar (see Figs. 16, 18) is due to the sharp decrease in the pair-breaking factor¹⁶⁰ in this range of pressure, which can be related to the enhancement under hydrostatic compression of Kondo screening of the magnetic moments of the cerium ions. Since the spins which are not entirely compensated hinder the appearance of superconductivity, the growth in T_c in the region $p = 0$ –5 kbars must be interpreted as the critical temperature T_c "tracking" the process of increasing T_K : $dT_K/dp \approx dT_{\text{max}}^{(1)}/dp \approx 0.7$ °K/kbar. This value is of the same order of magnitude as the derivative $dT_c/dp = 0.1$ –0.2 °K/kbar. The saturation of $T_c(p)$ for $p > 4$ –5 kbar apparently is due to the fact that in the range $p > 5$ kbar the Kondo spin screening is extremely effective, and the fully compensated magnetic ions no longer have any effect on T_c .

In Ref. 135 the influence of pressures up to 80 kbars on T_c was investigated for samples of CeCu_2Si_2 with excess copper, and it was observed that in the interval 25–40 kbars T_c increased up to 1.8 °K. It is not possible as yet to relate this large value of T_c with heavy-fermion superconductivity in any definite way, since in Ref. 135 neither dH_2/dT for $T = T_c$ nor the electronic contribution to the heat capacity were measured. Therefore, it is possible that the growth in T_c for p in the range 25–40 kbars is related to a structural transformation to a phase similar to LaRh_2Si_2 . We note that the compound LaRh_2Si_2 is a superconductor with $T = 3.8$ °K.¹⁶¹ The hypothesis that a structural transformation is possible is partially confirmed by the change in the character of the superconducting transition: whereas in the range $p < 19$ kbars the width of the transition is very small ($\Delta T_c = 0.04$ –0.08 °K), for $p = 25$ –40 kbars $\Delta T_c \approx 1$ °K.¹³⁵

The superconducting characteristics of $\text{CeCu}_{2+\delta}\text{Si}_2$

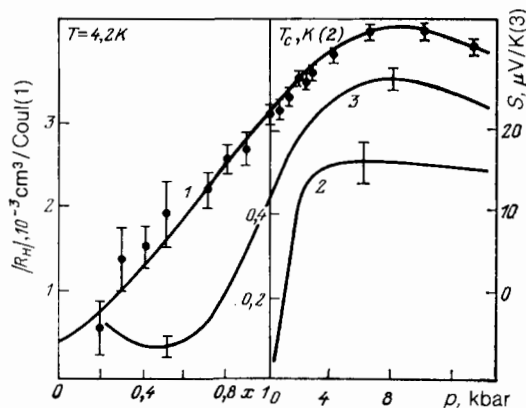


FIG. 18. (1) Variation in the Hall coefficient R_H ($T = 4.2$ °K); (3) the value of the thermoelectric power coefficient at the low-temperature extremum (minimum or maximum; see Fig. 6) and (2) the superconducting transition temperature T_c , in the series $\text{Ce}_x\text{La}_{1-x}\text{Cu}_2\text{Si}_2$, and in CeCu_2Si_2 under pressure.^{16,17}

are very sensitive to the deviation δ from stoichiometry.^{153,162,163} For example, a variation of $\delta \sim 1\%$ for copper decreases T_c from 0.67 °K to $T_c < 0.07$ °K. For copper, the dependence $T_c(\delta)$ appears to be the same as the pressure variation of the critical temperature $T_c(p)$ (see Figs. 16, 18). Variation of the copper content affects the residual resistance: $\rho_0 = 3-10 \mu\Omega \cdot \text{cm}$ for $\delta = +(1-3)\%$, while $\rho_0 = 50-80 \mu\Omega \cdot \text{cm}$ for $\delta = -(1-3)\%$. Replacing Ce, Cu, Si in CeCu_2Si_2 by other elements produces analogous variation in the degree of departure from stoichiometry. In studying the substitutional solid solutions $\text{Ce}_{1-x}\text{M}_x\text{Cu}_2\text{Si}_2$ ($\text{M} = \text{La, Y, Sc}$), $\text{Ce}(\text{Cu}_{1-x}\text{T}_x)_2\text{Si}_2$ ($\text{T} = \text{Ag, Au, Mn, Ru, Rh, Pd}$) and $\text{CeCu}_2(\text{Si}_x\text{Ge}_{1-x})_2$ it was established that in all cases substitution of an original element by another element led to a decrease in T_c . The critical concentration x_c corresponding to complete suppression of T_c varied in the range 0.5–10 at.%. For the alloys $\text{Ce}_{1-x}\text{M}_x\text{Cu}_2\text{Si}_2$ the value of x_c was larger the closer the lattice constant a in $\text{Ce}_{0.8}\text{M}_{0.2}\text{Cu}_2\text{Si}_2$ was to the value of a for CeCu_2Si_2 , i.e., the larger the chemical compression, the smaller the value of x_c . Thus, there is a definite correlation between x_c and T_K as estimated by the position $T_{\text{max}}^{(1)}$ of the maximum in $\rho(T)$.¹⁶²

In analyzing these data, it is necessary to take into account the following factors. First of all a decrease in the cerium content in CeCu_2Si_2 decreases the amplitude of the ASR. Secondly, according to the calculations of Tachiki and Maekawa,¹⁶⁴ NKLs in the coherent regime $T < T_{\text{coh}}$ exhibit a sharp decrease in the Coulomb repulsive energy between heavy fermions, which facilitates the appearance of superconductivity. Therefore the suppression of superconductivity in HFSs when impurities are introduced—both magnetic and nonmagnetic—can be treated as a consequence of disruption of coherence,^{16,17} since in view of the very narrow ~ 10 °K ASR, we need very small concentrations of impurities to bring about potential fluctuations $\Delta U \sim T_K \sim 10$ °K sufficient to destroy the coherent state. In this situation, the larger the difference in the parameter a between CeCu_2Si_2 and $\text{Ce}_{0.8}\text{M}_{0.2}\text{Cu}_2\text{Si}_2$, the smaller is the value of x_c needed to disrupt the coherence.

In the alloys $\text{Ce}(\text{Cu}_{1-x}\text{T}_x)\text{Si}_2$, in addition to the loss of coherence, there is also a shift in the d band relative to the $4f$ band, which can be quite large. This shift can occur when T is substituted for Cu since according to spectroscopic data⁹⁹ the overlap of the $4f$ and d bands varies strongly when Cu is replaced by Ag or Au. As opposed to the $3d$ band of Cu, the $4d$ band (Ag) and $5d$ band (Au) overlap very weakly with the $4f$ band. Therefore when Ag and Au are introduced it is possible for J and T_K to decrease, leading in turn to a decrease in T_c . In the alloys $\text{CeCu}_2(\text{Si}_{1-x}\text{Ge}_x)_2$ replacement of silicon by germanium causes an increase in the volume of the unit cell,¹⁶² which is analogous to application of “negative” pressure which decreases T_c .

In investigating single-crystal CeCu_2Si_2 obtained from slow cooling of CeCu_2Si_2 dissolved in an In or Sn melt, it has been observed¹⁵⁷ that replacement of Cu by In or Sn is analogous to shifting the Fermi level upward relative to the $4f$ level, i.e., In and Sn act as donor impurities. In copper-defi-

cient samples (i.e., those with an excess of indium or tin) the value of $E_F - E_{4f}$ grows, which leads to a decrease in J and T_K [see (1.4) and (1.1)]. Therefore, such samples must be located on the $T-x$ phase diagram to the left of the stoichiometric composition CeCu_2Si_2 . In these samples, as the temperature is decreased, the partially screened local magnetic moments of the Ce^{3+} ions are frozen out at $T_M = 3.5$ °K (see Fig. 16), and the superconducting transition is absent while the maximum $T_{\text{max}}^{(1)}$ of the curve $\rho(T)$ corresponds to a temperature $T_K \sim 8$ °K. In samples with excess copper (with insufficient In or Sn), on the contrary, the magnetic transition is absent, $T_c \approx 0.5$ °K and the value of $T_{\text{max}}^{(1)} \sim T_K$ rises to 14 °K.

In summary, the peculiarities in the behavior of CeCu_2Si_2 mentioned above when the composition and applied pressure are varied certainly underline the unusual nature of superconductivity in CeCu_2Si_2 ; these peculiarities are connected with the presence of the narrow ASR, which is very “sensitive” to the influence of various external parameters.

4.3. Heavy-fermion superconductivity in compounds of uranium and cerium

From 1979 to 1983, the compound CeCu_2Si_2 remained the only example of a superconducting HFS. In 1983, two new and unusual superconductors were discovered— UBe_{13} and U_6Fe ,¹⁶⁵ followed in 1984 by UPt_3 .^{6,10} In these compounds based on the $5f$ element uranium, characteristic anomalies occur analogous to those which were observed earlier in the NKL CeCu_2Si_2 (see Table III): a giant electronic specific heat coefficient, enhanced Pauli paramagnetism, a giant derivative of the upper critical field which correlated with the large value of $\rho_0\gamma$ (4.1), a strong dependence of T_c on magnetic and nonmagnetic doping, etc. It must be noted that unusual superconducting properties are most evident in the three HFS CeCu_2Si_2 , UBe_{13} and UPt_3 ; however, there are still other cerium and uranium superconductors (Tables IV, V) which occupy an intermediate position between heavy-fermion and normal superconductors.

To elucidate the nature of superconductivity in cerium compounds, it is of interest to compare their critical temperatures T_c^{Ce} (Table IV) with the critical temperatures T_c^{La} of the corresponding lanthanum homologues. Fulfillment of the inequality $T_c^{\text{Ce}} \gg T_c^{\text{La}}$ (e.g., for CeCu_2Si_2 and LaCu_2Si_2) can be considered a property which is intrinsic to nontrivial HFS superconductivity. It is possible that CeRu_2 (see Table IV) is also an unusual superconductor from this point of view.¹⁷⁰

For the uranium superconductors (Table V), there is a noteworthy correlation between γ and the $U-U$ spacing between uranium atoms: the larger the spacing between uranium atoms, the larger the low-temperature density of states at the Fermi level. It can be assumed that the above-mentioned regularity reflects the inverse proportionality of the dependence of γ on T_K (3.16), which is fulfilled particularly well for the cerium systems (see Table IV).

There is an analogy between the HFS superconductors CeCu_2Si_2 and UBe_{13} as regards the temperature dependence

TABLE IV. Normal and superconducting characteristics of cerium compounds¹⁶⁶; T_c^{Ce} , T_c^{La} are temperatures of the superconducting transitions of cerium and lanthanum homologues.

Compound	χ_p CGS/mole	γ mJ/mole K ²	T_K , K	T_c^{Ce} , K	T_c^{La} , K
CeRu ₂ ¹⁶⁷⁻¹⁷⁰	$0.6 \cdot 10^{-3}$	23	770	6.2	4.1
CeRu ₂ Si ₂ ¹⁶⁹	$0.9 \cdot 10^{-3}$	39	440	1.0	7.2
CeSn ₂ ^{169, 171, 172}	$1.5 \cdot 10^{-3}$	53	270	< 0.02	6.4
CeCu ₂ Si ₂ ¹⁵⁵	$6.5 \cdot 10^{-3}$	1000	10	0.65	< 0.02

of the specific resistivity $\rho(T)$. The functions $\rho(T)$ have two different regions of Kondo growth of ρ , characteristic of the splitting of the f level in the crystal field, i.e., $\Delta_{\text{CF}} > T_K$. In addition, the functions $\chi(T)$, $C(T)$, and $R_H(T)$ are very similar for the two compounds over a wide temperature range. Based on the analogy between the low-temperature behavior of CeCu₂Si₂ and UBe₁₃, it can be assumed that UBe₁₃ also exhibits a narrow peak in the density of states at E_F —that is, an ASR; i.e., UBe₁₃ too is apparently a “ $j = 1/2$ ” NKL with $E_R = E_F$.

With regard to the set of its low-temperature properties, UPt₃ is strikingly different from CeCu₂Si₂ and UBe₁₃. First of all, in UPt₃ the Kondo growth in ρ is absent, and over the whole range of temperatures $T < 300$ °K the behavior of $\rho(T)$ is metallic.¹⁰ In the specific heat $C(T)$ of UPt₃ there is a contribution proportional to $T^3 \ln(T/T_{sf}^*)$, which is characteristic of a contribution to $C(T)$ from spin fluctuations.¹⁰ On the whole, the temperature dependences of $C(T)$, $\rho(T)$ and $S(T)$ in UPt₃ are very similar to the corresponding dependences in the magnetic spin-fluctuation compound UAl₂.^{178,179} If we do not include the paramagnetic contribution to the magnetic susceptibility and electronic specific heat coefficient, the Pauli paramagnetism χ and γ are proportional to the density of states at the Fermi level. In connection with this, it is interesting to compare the ratio χ/γ for the three HFS—CeCu₂Si₂, UBe₁₃ and UPt₃. In the first two compounds, χ is enhanced by a smaller factor than γ ; this is particularly noticeable in CeCu₂Si₂. In UPt₃, on the contrary, χ is enhanced almost four times as much as γ , i.e., in UPt₃ there is an appreciable paramagnon contribution to

χ ; thus, we cannot exclude the possibility of coupling between heavy fermions via paramagnons.¹⁸⁰

The NKLs CeCu₂Si₂ and UBe₁₃ are characterized by negative magnetoresistance, while UPt₃ has a rather large positive magnetoresistance. In a field $H \approx 90$ kOe at $T = 1.2$ °K, the relative change in magnetoresistance for these three compounds amounts to 4.5%, 34% and 41% respectively.¹⁸¹

The comparison we have presented of the HFS superconductors CeCu₂Si₂, UBe₁₃, and UPt₃ shows that UPt₃ is a compound in which spin fluctuations make a large contribution to the various low-temperature characteristics. Such compounds seem to prefer ferromagnetic pairing; based on this point of view, we conclude that of all the well-known HFS superconductors UPt₃ is the one in which triplet pairing is most likely to occur.

4.4. On the nature of the unusual superconductivity of heavy-fermion systems

In a normal BCS superconductor, for $T < T_c$ a nonzero gap $\Delta(\mathbf{k})$ opens up for all directions of \mathbf{k} in the Brillouin zone. Therefore, in experiments which measure the electronic specific heat $C(T)$, the ultrasonic absorption $\alpha(T)$, the rate $1/T_1$ of spin-lattice relaxation measured by NMR and the electronic contribution to the thermal conductivity $K_e(T)$, an obvious exponential dependence appears in the temperature dependence of $C(T)$, $\alpha(T)$, $1/T_1(T)$ and $K_e(T)$ below the critical temperature T_c . In all HFS superconductors, instead of the usual exponential dependence below T_c , these parameters exhibit a power-law

TABLE V. Physical properties of uranium superconductors.⁹⁷

Compound	T_c , °K	$-dH_{c2}/dT$, ($T = T_c$), kOe/°K	γ mJ/mole °K ²	χ , 10^{-4} CGS/mole	Spacing between uranium atoms, Å
UBe ₁₃ ^{7,173-176}	0.8–0.95	257–440	1100	151	5.13
UPt ₃ ^{10, 176, 177}	0.5	63 (H c) 40 (H ⊥ c)	450	89	4.1
U ₂ PtC ₂ ⁹⁷	1.47	90	75 *	15–22 *	3.52
U ₄ Fe ¹⁶⁶	3.8	34	25 *	5.0 *	3.2
α-U ⁹⁷	2.1	?	12	3.7	3.12

*The values of γ and χ are calculated per uranium atom for convenience in comparison with the rest of the compounds, which contain one atom of U or Ce.

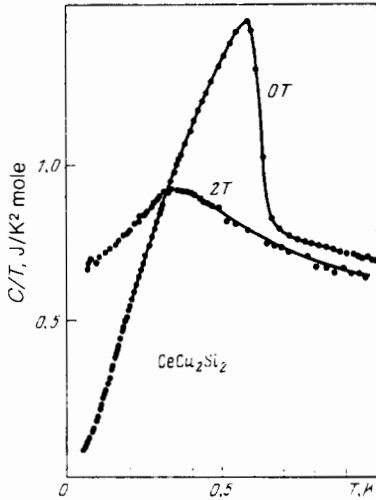


FIG. 19. Temperature dependences of the specific heat for CeCu_2Si_2 for $H = 0$ and $H = 20$ kOe.¹⁷⁸ The reduction of the superconducting gap to zero follows a straight line for $T < T_c$ if we use the coordinates C/T and T .

temperature dependence (Figs. 19, 20); for CeCu_2Si_2 , $C(T) \sim T^2 (T < T_c)$, and $C(T) \sim T^3 (T \ll T_c)$ ¹¹⁸; $1/T_1 \sim T^3$,^{182,183} $K_c(T) \sim T^2 (T \ll T_c)$ ^{118,178}; in UBe_{13} , $C(T) \sim T^3$,¹⁸⁴ $K_c(T) \sim T^2 (T \ll T_c)$,¹⁷⁸ $1/T_1 \sim T^3$ ¹⁸⁵; in UPt_3 , $C(T) \sim T^{2-3}$,^{10,178} $K_c(T) \sim T^{2,178}$ and $\alpha(T) \sim T^2$.^{15,128} The absence of an exponential dependence is a sign of a *vanishing superconducting gap* $\Delta(\mathbf{k})$ along certain directions of the vector \mathbf{k} . Let us recall that vanishing of the

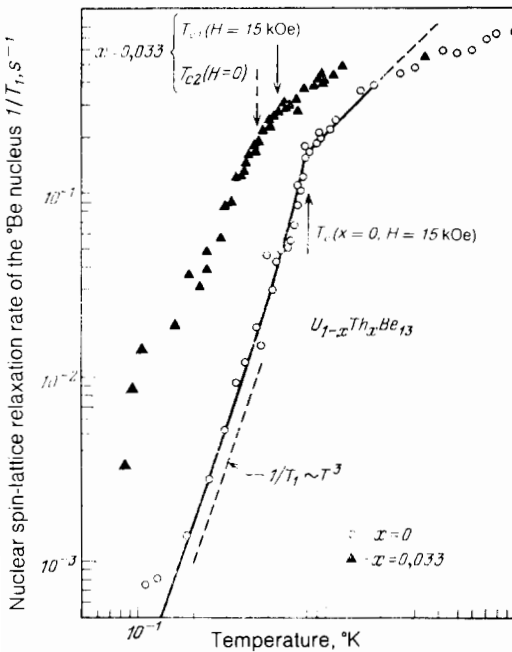


FIG. 20. Temperature dependences of the rate $1/T_1$ of spin-lattice relaxation of ^9Be in $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ ($x = 0, x = 0.033$).¹⁸⁵ The arrows denote the location of the superconducting transition temperature in a field $H = 15$ kOe. The dependence $1/T_1 \sim T^3$ corresponds to the superconducting gap $\Delta(\mathbf{k})$ going to zero along some curve on the Fermi surface.

gap $\Delta(\mathbf{k})$ at certain *points* of the Fermi surface leads to the following power-law dependences for $T \leq T_c$: $C(T) \sim T^3$, $\alpha(T) \sim T^4$, $1/T_1 \sim T^5$, $K_c(T) \sim T^3$; while the vanishing of the gap along some curve on the Fermi surface must be accompanied by the existence for $T \leq T_c$ of power laws of the form $C(T) \sim T^2$, $\alpha(T) \sim T^2$, $1/T_1 \sim T^3$, $K_c(T) \sim T^2$.

The vanishing of the superconducting gap in the HFS CeCu_2Si_2 , UBe_{13} , and UPt_3 points to the unusual character of superconductivity in these compounds. In systems with strong electron-electron correlations (including all the HFS) which contain f electrons with a Hubbard repulsion of $U \sim 8$ eV, in place of the usual BCS coupling of two s electrons with opposite spins it turns out to be more advantageous to couple p or d electrons, i.e., electrons with nonzero orbital angular momentum $l \neq 0$; this includes the cases of singlet ($S = 0$) and triplet ($S = 1$) superconductivity.

If we ignore the real crystal structure and assume that the heavy fermions form an isotropic Fermi liquid, in order to describe the superconducting properties of isotropic systems with p -pairing we can use results obtained for liquid ^3He .^{187,188} In addition to phase A (the Anderson-Brinkman-Morrel (ABM) phase) and phase B (the isotropic Balian-Werthamer (BW) phase) which appear in ^3He , there is also a polar (P) phase and a planar (PL) phase which correspond to local minima of the free energy. The squared moduli of the superconducting gaps for all these phases are given by the relations $\Delta_{\text{ABM}}^2 (1 - K_z^2)$, Δ_{BW}^2 , $\Delta_{\text{P}}^2 k_z^2$ and $\Delta_{\text{PL}}^2 (1 - k_z^2)$ respectively. It is clear that for the polar phase the superconducting gap vanishes along a curve passing through the equator, while for the ABM and planar phases the superconducting gap reduces to zero at two points ($k_z = \pm 1$) corresponding to the poles.

Thus, on the basis of a comparison with the superfluid phases of ^3He , we can conclude that the HFS superconductors UPt_3 and CeCu_2Si_2 "resemble" the polar phase, since for these compounds the character of the variations of $C(T)$, $\alpha(T)$, $K_c(T)$ and $1/T_1(T)$ points to the vanishing of the superconducting gap along a curve for $T \leq T_c$. However, such a comparison cannot be entirely correct, since for all well-known HFS superconductors, as opposed to ^3He , the spin-orbit interaction is very strong, and consequently the spin is not a good quantum number, so that in the majority of cases the total magnetic moment j is "operative," this vector "catches" on lattice modes and rotates with the lattice as the symmetry changes.^{11,189} This circumstance allows us to conduct a symmetry-based analysis of the possible superconducting classes for systems with cubic (UBe_{13}), hexagonal (UPt_3) and tetragonal (CeCu_2Si_2) lattices.^{11,189-191} Gor'kov has shown¹⁹⁰ that in order to identify the superconducting classes which can occur in HFS superconductors data can be used from the angular dependence of the upper critical field H_{c2} obtained by rotating the vector \mathbf{H} perpendicular to the high-symmetry axes. For triplet superconductors with $S = 1$, in the case of tetragonal symmetry (CeCu_2Si_2) the presence of significant anisotropy of the fields H_{c2} is predicted with a fourfold symmetry axis for $T_c - T \ll T_c$,¹⁹⁰ and also the possibility of a spontaneous magnetic moment appearing in a superconducting transi-

tion¹⁸⁹ (even in the absence of an external field).

The anisotropy of H_{c2} has been investigated in Ref. 192 for CeCu_2Si_2 near T_c —to be precise, the anisotropy of the derivative— $dH_{c2}/dT(T = T_c)$. Single crystals were studied in the form of films perpendicular to a plane which coincides with the tetragonal axis c . When the vector \mathbf{H} intersects the plane of the sample $\mathbf{H} \perp c$, the derivative of the upper critical field increases by a factor 1.6–1.7 compared to $\mathbf{H} \parallel c$; unfortunately, it is not yet clear whether this is connected with the coincidence of the plane in which the vector \mathbf{H} is rotating and the null-line $\Delta(\mathbf{k}) = 0$ located at the equator, or with “geometric” factors—the sample shape, surface superconductivity, precipitation of impurities, etc. Doubtless additional investigations will be required to answer these questions.

The authors of Ref. 193, while investigating the angular dependence $H_{c2}(\varphi)$ in CeCu_2Si_2 near T_c in a basal plane $\mathbf{H} \perp c$, observed no anisotropy of H_{c2} within $\pm 7\%$ limits, although a tetragonal anisotropy in H_{c2} was found; this latter turns out to be dependent on the material of the crucible in which the single crystals were grown: $\sim 40\%$ for a tungsten crucible and $\sim 10\%$ for a boron nitride crucible. The causes of such behavior of $H_{c2}(\varphi)$ are not yet clear.

The first measurements of anisotropy of H_{c2} in UBe_{13} were performed by Alekseevskii and coauthors.¹⁹⁴ For two orientations— $\mathbf{H} \parallel c_2$ and $\mathbf{H} \parallel c_4$ —the same values of the upper critical field are obtained near T_c , i.e., for these directions of \mathbf{H} there is no anisotropy in $-dH_{c2}/dT(T = T_c)$.

In concluding this section, we note that at present investigations of HFS superconductivity are in a preliminary phase, and there are many unresolved questions in the area. However, it is already clear that HFS superconductivity is unusual and nontrivial. The nature of the attraction between heavy fermions is not clear; this attraction could be due to a phonon mechanism, in which case it is necessary to take into account Kondo-collapse effects in the NKLs UBe_{13} and CeCu_2Si_2 ,¹⁵⁹ although the possibility of an attraction due to the exchange interaction mechanism has also been predicted.¹⁴ The superconducting state itself can be either a singlet or a triplet. The unusually high density of states $g(E)$ at the Fermi level for CeCu_2Si_2 and UBe_{13} , as was discussed above, is related to the formation of a giant ASR, i.e., CeCu_2Si_2 and UBe_{13} are superconducting NKLs. The compound UPt_3 differs significantly from CeCu_2Si_2 and UBe_{13} , and it is not fully understood how the enhancement of $g(E)$ comes about in this material.

The unusual superconductivity of HFS systems has attracted much interest recently, and is being studied with extraordinary intensity. For this reason, the material presented in the present review cannot lay claim either to completeness or conclusiveness; in fact, it serves only as an introduction to the physics of superconducting systems with heavy fermions.

5. CONCLUSION

The basic aim of the present review was an analysis of the anomalous low-temperature properties of NKLs. In this review we have shown that the existence of these anomalies

is connected with the formation at the Fermi level of an exceedingly narrow ($\sim 10^\circ\text{K}$) high-amplitude ASR.

On the basis of the set of their low-temperature properties the NKLs have no analogues among all the known types of solids, and in fact must actually be studied as a new class of metallic systems.

The treatment presented in this review of many cerium IV compounds as CKS with $T_K > \Delta_{CF}$ and $E_R > E_F$ leads to a different interpretation of the low-temperature properties of these systems, and will stimulate the design of new experiments in which the transition from $T_K \ll \Delta_{CF}$ to $T_K > \Delta_{CF}$ can be induced with the aid of such external influences as pressure and controlled variation of composition.

Various experiments in which one could track directly by spectroscopic means the progressive appearance of the ASR as the temperature is lowered from $T \gg T_K$ to $T \lesssim T_K$, as well as experiments on studying the nature of the renormalization of the Fermi surface of NKLs in the coherent regime when the heavy-fermion bands have already been formed also appear to be of great interest.

One of the central issues in the physics of NKLs is indisputably the need for further experimental and theoretical investigation of superconductivity in heavy-fermion systems where in principle there can occur pairing of electrons with nonzero orbital angular momentum.

It should be pointed out here that despite the enormous interest related to *superconductivity* of heavy fermion systems, an understanding of all the peculiarities of this astonishing phenomenon can be achieved only through actual understanding of the properties of HFS in the *normal phase* for $T > T_c$. This circumstance was repeatedly emphasized in the concluding words of C. Varma¹⁹⁵ at the IVth International Conference on Valence Fluctuations.

One can proceed further along the chain of transitions: Kondo-impurity to concentrated (“dense”) Kondo systems, to the “superdense” Kondo systems CeSb , CeBi .¹⁹⁶ Whereas in systems with Kondo impurities a situation occurs when many conduction electrons are associated with each Kondo center, $n_m \gg 1$, while in CKS this number is close to unity $n_m \sim 1$, in the “superdense” Kondo systems we have the opposite relation $n_m \ll 1$. Investigation of such systems, in which the value of n_m can be varied over wide limits, also appears to be very promising.

Areas such as the physics of magnetic Kondo lattices— CeB_6 , CeIn_3 , CeAl_2 , etc.—touch directly on the physics of nonmagnetic CKS. Work in this area is extremely important to the understanding of the sequence of phenomena in the progression from magnetic metals ($T_{RKKY} \gg T_K$) to magnetic Kondo lattices ($T_{RKKY} \gtrsim T_K$) to nonmagnetic Kondo lattices ($T_{RKKY} \ll T_K$); however, due to the limited scope of the present review, we have not been able to dwell on this interesting question.

* * *

We completed work on this manuscript in June of 1985; however, the study of NKLs, and primarily of heavy-fermion superconductors, is developing so vigorously that during the time our manuscript was being prepared for publication a large number of papers has appeared in which

extremely interesting data have been obtained.

It is sufficient to say that the problem of the behavior of heavy fermions in metals received primary attention at past international conferences on crystal fields in anomalous f -systems²⁰⁷ (Sendai, Japan, April 1985), on the theory of heavy-fermion systems²⁰⁸ (the VIIIth Taniguchi symposium, Japan, April 1985), on magnetism²⁰⁹ (San Francisco, USA, August 1985) and on superconducting materials and mechanisms of superconductivity (Iowa, USA, May 1985). In the course of the next half-a-year, international conferences are imminent on rare earths (Hamilton, Canada, June 1986), on anomalous systems based on rare earths and actinides (Grenoble, France, June 1986) and on valence fluctuations (Bangalore, India, January 1987).

In connection with this, it behooves us to note certain papers which have recently appeared which reflect progress in the physics of Kondo lattices and HFS. First of all, two new and unusual heavy-fermion superconductors have been discovered—CePb₃²⁰⁵ and URu₂Si₂.²⁰⁶ The compound CePb₃ is found to be a magnetic Kondo lattice, in which for sufficiently low temperatures a strong magnetic field induces first a transition to a nonmagnetic and then (at ~ 120 kOe) to a superconducting state. The magnitude of the magnetic field which destroys the induced superconductivity has not yet been determined.

In the compound URu₂Si₂, in measuring the magnetic susceptibility, specific heat and specific resistance a magnetic transition has been observed²⁰⁶ at $T_M = 17.5$ °K (preceding the superconducting transition at $T_c = 0.8$ °K), while the slope of the derivative $dH_{c2}/dT(T = T_c) = 40$ kOe/°K correlates with a high value of the coefficient of the electronic specific heat $\gamma = 180$ mJ/mole °K. This correlation points to the possibility that heavy-fermion superconductivity is accompanied by antiferromagnetic ordering.

Much interest attaches to ongoing investigation of the nature of a second peak (in the temperature region below T_c) in the specific heat of $U_xTh_{1-x}Be_{13}$.²¹⁰ It turns out that the second peak corresponds to a magnetic transition with strongly-reduced uranium magnetic moments. Apparently, the magnetic transition in $U_xTh_{1-x}Be_{13}$, like that in $Ce_xLa_{1-x}Cu_2Si_2$ (see Fig. 16) corresponds to an intersection of the branches $T_c(J)$ and $T_M(J)$ as the parameter J is varied in the CKS.

Further study of the anisotropy in H_{c2} for CeCu₂Si₂²¹¹ has made it possible to exclude geometric factors. Thus, it has been established that the derivative $dH_{c2}/dT(T = T_c)$ is isotropic (to $\sim 8\%$ accuracy) in the basal plane, and sharply decreases by a factor of 1.5–2 within an angular interval of $\pm 5^\circ$ when the vector H deviates from this plane. Such anisotropy in H_{c2} can correspond either to anisotropic s -pairing²⁰⁸ or p -pairing,²¹² but with an effective mass along the C_4 axis which would exceed the basal-plane effective mass by not less than a factor of 2 times. In the latter case, according to Ref. 212 the anisotropy of H_{c2} in the basal plane must be very small.

The following empirical regularity is worth noting: in CeCu₂Si₂,²¹¹ UPt₃¹⁷⁷ and URu₂Si₂²⁰⁶ the value of H_{c2} is larger along those directions along which the magnetic susceptibility is smaller.

An anomalous ultrasonic absorption has been observed in the compound UBe₁₃ for $T < T_c$, pointing to the possibility of collective modes appearing, analogous to those which exist in superfluid ³He. The authors of Ref. 213 suggest that this circumstance indicates positive evidence in favor of triplet pairing in UBe₁₃.

Interesting data have been obtained in investigating the Hall coefficient $R_H(T)$ in the NKLS CeCu₆,^{200,201} UBe₁₃,²⁰³ CeAl₃,²⁰² and CePd₃.²⁰⁴; a transition into the temperature range $T < T_{coh}$ which corresponds to the establishment of coherent Kondo spin fluctuations is accompanied by a sharp decrease in $R_H(T)$ and a change in the nature of the field dependences of the Hall voltage.^{200,201,203}

In limiting ourselves to the examples cited above, we want to point out that additional references on the problem of HFS and CKS can also be found in Ref. 214 and Refs. 208 and 215, in which recent results are analyzed which have been obtained in the theory of HFS and CKS. The authors of the present review would like to stipulate beforehand that in view of the objective circumstances connected with the remarkably rapid growth in the volume of information on HFS and CKS, the bibliography presented in this review can have no claims to completeness.

The authors express sincere acknowledgment to A. A. Abrikosov, N. E. Alekseevskii, L. P. Gor'kov, D. I. Khomskii, A. I. Buzdin, and F. G. Aliev for useful discussions, and to F. Steglich, J. Smith, T. Kasuya, A. Okiji, D. Wohlleben, S. Maekawa, T. Komatsubara, and R. Parks for sending preprints.

¹In this article we use the following abbreviations: CKS—concentrated Kondo system, NKL—nonmagnetic Kondo lattice, HFS—heavy fermion system, ASR—Abrikosov-Suhl resonance, IVC—intermediate valence compound.

²We note that for a typical concentrated Kondo system with $T_K \sim 10$ °K, for all ranges of temperature—from $T \sim 300$ °K down to $T_{coh} \sim T_K/10$ (see below)—the ASR amplitude can be considered to be independent of temperature.^{34,35,198}

³Here and from now on, we will understand J to mean the modulus of the exchange interaction constant.

⁴This slope cannot be interpreted as a growth in the ASR,¹⁹⁸ which has already formed for $T \sim (10-100)T_K \sim 300$ K. The increase in C/T as $T \rightarrow 0$ is connected with a decrease in the width of the derivative of the Fermi distribution down to values smaller than the ASR width.

⁵The transition to the coherent regime also sharply decreases the Hall coefficient $R_H(T)$ for the NKLS CeCu₆,²⁰⁰⁻²⁰¹ CeAl₃,²⁰² UBe₁₃,²⁰³ while for CeCu₆ and UBe₁₃ $R_H(T)$ even changes sign. The destruction of periodicity of the Kondo centers significantly weakens this fall-off in $R_H(T)$.^{202,204}

⁶Recently, two new superconducting HFS have been discovered: in the magnetic Kondo-lattice CePb₃,²⁰⁵ superconductivity is induced by strong magnetic fields $H > 120$ kOe; in the HFS URu₂Si₂²⁰⁶ with $\gamma = 180$ mJ/mole K², the magnetic transition ($T_M = 17.5$ °K) is a precursor to superconductivity ($T_c = 0.8$ °K).

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Translated by Frank J. Crowne