A new Kondo lattice $CeSi_{2-x}Ga_x$

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Substitution of Ga for Si in $CeSi_{2-x}Ga_x$ leads to a suppression of the magnetic ground state of Ce^{3+} ions and at $x > 1$ to a change to conditions corresponding to a nonmagnetic Kondo lattice with heavy Fermi electrons which are characterized by a huge coefficient $\gamma \sim 1800$ mJ/(mole·K^2).

By varying the composition of the Kondo lattice it is possible to change the ratio between the Kondo temperature T_K and the temperature T_{RKKY} which characterizes the center-to-center interaction of the spins through the spin–density oscillations. The temperatures T_K and T_{RKKY} depend on the exchange–interaction parameter J in different ways: $T_K \sim E_F \exp[-1/(Jg(E_F))]$, where $g(E_F)$ is the density of the electronic states at the Fermi level, $T_{RKKY} \sim J^2/W$, and W is the band width. This possibility is made use of to cover the entire range of states between the magnetic Kondo

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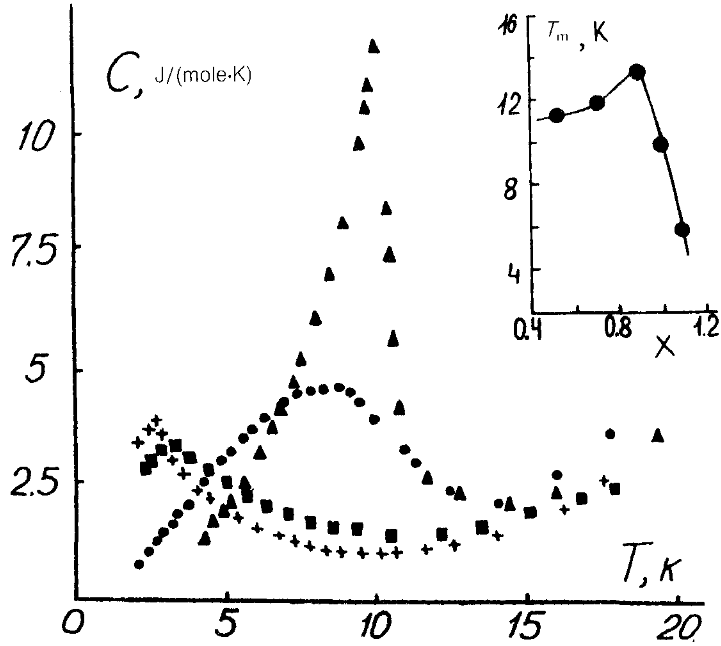

FIG. 1. Temperature dependences of the specific heat $C(T)$ of $CeSi_{2-x}Ga_x$ (▲—$x = 0.7$; ●—$x = 1.0$; + —$x = 1.2$; ■—$x = 1.3$). Inset—Concentration dependence of the magnetic-order temperature.

lattice ($T_K \leqslant T_{RKKY}$) and the nonmagnetic Kondo lattice ($T_K \geqslant T_{RKKY}$). The electronic contribution to the specific heat γT for nonmagnetic Kondo lattices should increase as the given compound approaches the critical region $J = J_c$, since $\gamma \sim 1/T_K$ in nonmagnetic Kondo lattices[1] and T_K decreases as $J = J_c$ is approached from the right (see the inset in Fig. 2).

The empirical method of obtaining nonmagnetic Kondo lattice with a surface-enhanced (giant) Abirkosov-Sula resonance, which we discussed above, $\gamma \sim g^R (E_F)$, is the principle upon which the search for new systems with heavy fermions is based.

In our study we have conducted this search by varying the composition of the system $CeSi_{2-x}Ga_x$. We used polycrystalline samples with $x = 0.7$–1.3 to measure the temperature dependence of the specific heat $C(T)$ and the resistivity $\rho(T)$. At $T \sim 10$ K the system $CeSi_{2-x}Ga_x$ acquires, in the single-phase region of gallium concentrations $x > 0.5$ (Ref. 2), a ferromagnetic order at reduced local magnetic moments of Ce^{3+} due to the Kondo effect.[3] With an increase in the gallium concentration from $x = 0.9$ to $x = 1.3$, the magnetic transition becomes progressively more diffuse, while the magnetic-transition temperature (the inset in Fig. 1) and the magnetic-moment saturation decrease rapidly.

At $T = 10.5$ K the specific heat of the compound $CeSi_{1.3}Ga_{0.7}$ (Fig. 1) behaves anomalously during the ferromagnetic transition. At $T = 8$–9 K the $C(T)$ peak for the composition with $x = 1$ is broadened appreciably. The presence of a magnetic transition makes it more difficult to determine the value of γ. Extrapolation of high-temperature regions, $T > 10$ K, on the C/T axes from T^2 yields the value $\gamma_{\text{extrap}} \sim 100$ mJ/(mole·K^2), which is approximately the same for all compositions with $x = 0.7$–1.3.

At $x = 1.2$ and 1.3, the shape of the $C(T)$ curves is characteristic of nonmagnetic Kondo lattices with a low Kondo temperature,[1] in which $T_K < \Delta_{CF}$, where Δ_{CF} is the f-level splitting in a crystalline field. If the lowest state which is split off by the crystal field is a doublet $j = 1/2$, then the Abrikosov-Sula resonance is exactly at the Fermi level ($E^R = E_F$), since the degree of filling of the resonance at $T < T_K$ Δ_{CF} is determined by the degeneracy of the lowest state that has been split off. The low-temperature maxima on the $C(T)$ curve are not related to the magnetic order, whose absence was indicated by the measurements of the magnetization up to 1.5 K. The specific heat[4] calculated in the Coqblin-Schrieffer model by the Wiegmann-Andrew method on the basis of the equation

$$C(T) = 2jk_B \int_{-\infty}^{+\infty} \frac{g^R(E)(E/(2k_B T))^2}{\cosh^2(E/(2k_B T))}\, dE, \quad (1)$$

where the shape of the Abrikosov-Sula resonance near $E_F g^R(E)$ is approximated by a Lorentzian, is in good agreement with the experimental curves of $C(T)$ for $T_K^{\text{theor}} \approx 4$ K. The calculated and experimental (for $x = 1.3$) curves are shown in Fig. 2

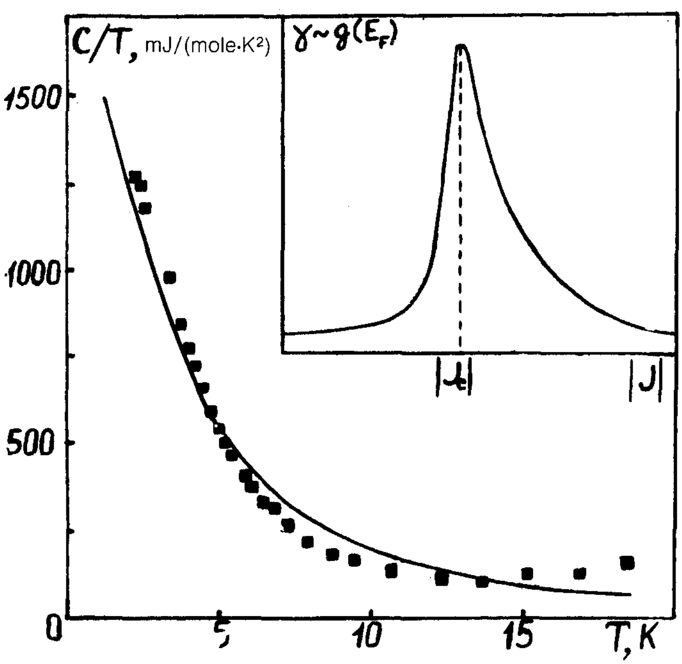

FIG. 2. Experimental temperature dependence and temperature dependence calculated from Eq. (1) C/T (solid line) of nonmagnetic Kondo lattice of $CeSi_{0.7}Ga_{1.3}$. The inset shows the electronic coefficient γ plotted as a function of the exchange interaction parameter J.

in $C/T(T)$ coordinates. As can be seen from Fig. 2, the value of the parameter $\gamma(0)$ is ~ 1800 mJ/(mole·K^2). Substituting this value of $\gamma(0)$ in the expression[5]

$$T_K^\gamma = \pi^2 R/(6\gamma) \qquad (2)$$

we find $T_K^\gamma \approx 3.3$ K, in nearly complete agreement with the value of T_K^{theor}, which was used as a parameter in Eq. (1). A plot of the Hall coefficient $R_H(T)$ for the compounds with $x > 1$, characteristic of nonmagnetic Kondo lattices, was previously observed by Brandt *et al.*[6]

By calculating the area under the curve of the specific heat with the $C/T(T)$ axes we were able to estimate the entropy associated with the low-temperature anomaly. For all tested compounds an increase in the entropy is approximately the same: $\Delta S \sim 4.69$ J/K $\approx R\ln 2$. Consequently, the magnetic level $j = 5/2$ of the Ce^{3+} ions is strongly split by the crystalline field, and the lowest energy state is the doublet. It can

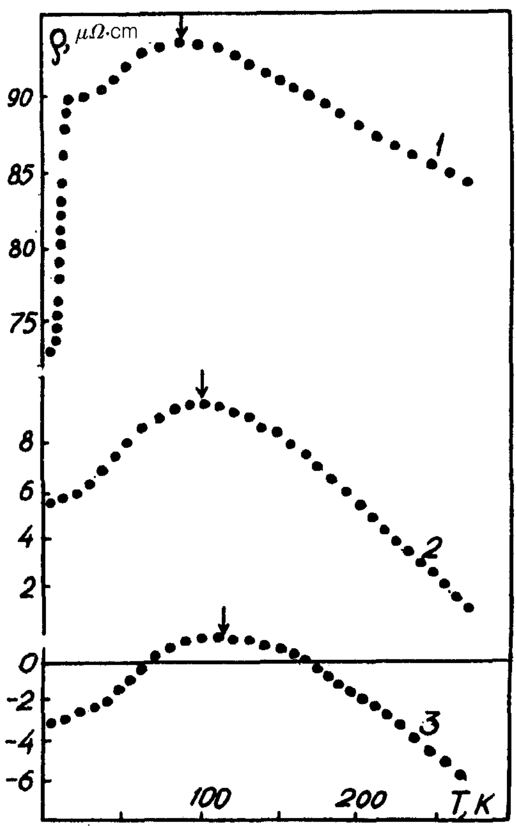

FIG. 3. Temperature dependences of the magnetic components of the resistivity $\rho_m = \rho(CeSi_{2-x}Ga_x) - \rho(LaSi_{2-x}Ga_x)$ [1—$x = 1$; 2—$x = 1.2$; 3—$x = 1.3$]. The temperature peaks T_{max} are indicated by arrows.

be assumed that the general arrangement of the levels is similar to the arrangement of the levels in a $CeSi_x$ Kondo lattice, whose crystal structure is the same as that of the $CeSi_{2-x}Ga_x$ system, with $\Delta_{CF1} = 160$ K and $\Delta_{CF2} = 330$ K (Ref. 7). In this case two characteristic Kondo temperatures must be introduced: T_K^{high} and T_K^{low} (Ref. 8):

$$T_K^h = (T_K^l \Delta_{CF1} \Delta_{CF2})^{1/3} . \quad (3)$$

The value of T_K^h can be estimated from the position of the maxima on the temperature dependences of the magnetic component $\rho_m(T)$, which was determined by subtracting the resistivity of the lanthanum compound $LaSi_{2-x}Ga_x$ from the resistivity of $CeSi_{2-x}Ga_x$. The temperature maximum T_{max} on the $\rho_m(T)$ curves increases with increasing gallium concentration (Fig. 3), and the value of T_{max} is in agreement with the value of T_K^h which is estimated from γ_{extrap}. Using Eq. (3), we find that $T_K^l \sim 10$ K, which is close to the values of T_K^γ and T_K^{theor}. Accordingly, an increase in the gallium concentration causes the exchange interaction parameter to increase to J_c, and $CeSi_{2-x}Ga_x$ undergoes a transition from the magnetic lattice to a nonmagnetic Kondo lattice. In the case of a magnetic composition with $x = 0.7$ $T_K^h \approx T_{max} \approx 93$ K, $T_K^l \approx 9\ K < T_m$ and the low-temperature Kondo spin fluctuations are suppressed due to the magnetic ordering of the Ce^{3+} moments. For CeSiGa $T_K^h \approx 106$ K, $T_K^l \approx 14$ K, and the relation $T_K^l \geqslant T_m$ is satisfied most exactly. This leads to a broadening of the magnetic transition because of the Kondo fluctuations of the magnetic moment. The compositions with $x = 1.2$–1.3 are nonmagnetic Kondo lattices ($T_K^h \approx 119$ K, $T_K^l \approx 19$ K) with $\gamma \sim 1800$ mJ/(mole·K^2), in which the presence of heavy fermions is associated with the generation of a giant Abrikosov-Sula resonance with $E^R = E_F$.

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