# Transition of a Kondo system CeNiSn to an insulating state at low temperatures

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The first stoichiometric Kondo lattice based on cerium, CeNiSn, has been detected. Below $T = 10$ K this lattice undergoes a transition to a coherent insulating state. The effect of hydrostatic pressure and of the change in the composition on the ground state of CeNiSn has been studied.

**1**. The most urgent problem in the physics of systems with heavy fermions today is the coherent state. The available experimental data show that there is a transition from a incoherent Kondo scattering regime at $T > T_K$ ($T_K$ is the typical Kondo temperature, $T_K \sim 10$ K) to a coherent regime at $T < T^* \approx T_K/5$ (Refs. 1 and 2). The appearance of a coherent state is manifested in the decrease of the "Fermi-liquid" electrical resistance[3] and in the maximum in the electronic part of the specific heat,[1] $\gamma$. This situation led to the assumption that a gap is formed in the electron state density along certain directions in the Brillouin zone. On the other hand, in several recently

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discovered compounds based on rare-earth elements, primarily Sm and Yb [$SmB_6$ (Ref. 4) and $YbB_{12}$ (Ref. 5), for example] each magnetic center appears to have exactly on electron[6] and the transition to the coherent state induces an exponential increase of the "Fermi-liquid" electrical resistivity, rather than its decrease. In the present letter we report the results of a multifaceted study of galvanomagnetic, thermoelectric, thermodynamic, and magnetic properties of the CeNiSn system, on the basis of which we have reached the conclusion that we have found the first stoichiometric Ce compond in which a coherent gap, similar to that observed in $SmB_6$ and $YbB_{12}$, is formed in the electronic state density. Takabatake *et al.*[7] and Aliev *et al.*[8] reported earlier in a short communication that a coherent dielectric state can exist in CeNiSn. This assertion was based soley on resistive measurements.

2. The procedure for galvanomagnetic and thermoelectric measurements was described in detail by Aliev *et al.*[9] The heat capacity was measured over the temperature range $2 < T < 15$ K, using a quasiadiabatic pulsed method. The polycrystalline CeNiSn samples were synthesized by fusing a starting stock comprised of pure components containing at least 99.99% basic component. The fusion was carried out in an electric-arc furnace in purified argon and the samples were then homogenized at a temperature of 600 °C. The crystal symmetry of the CeNiSn compound corresponds to the structural type described in Ref. 10.

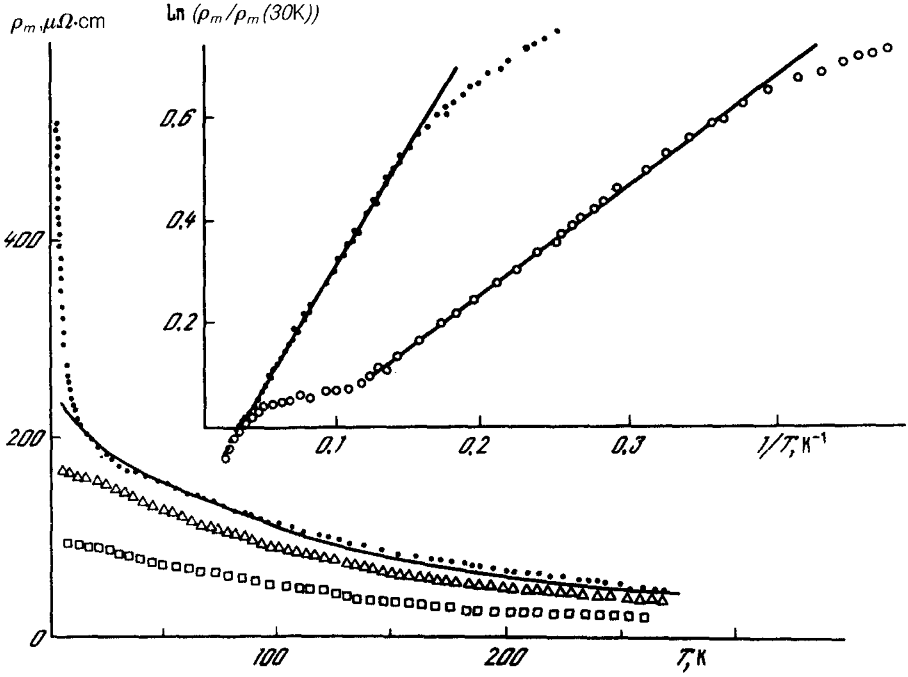


FIG. 1. Temperature dependence of the magnetic component of the resistivity of $Ce_{1-x}La_xNiSn$ for the compositions with $x = 0$ (points); $x = 0.1$ (line); $x = 0.5$ (Δ); $x = 0.2$ (□). The inset shows the curves for $\mathrm{Ln}[\rho_M/\rho_M(30\ \mathrm{K})] = f(1/T)$ for sample 58 (●) and sample 52 (○).

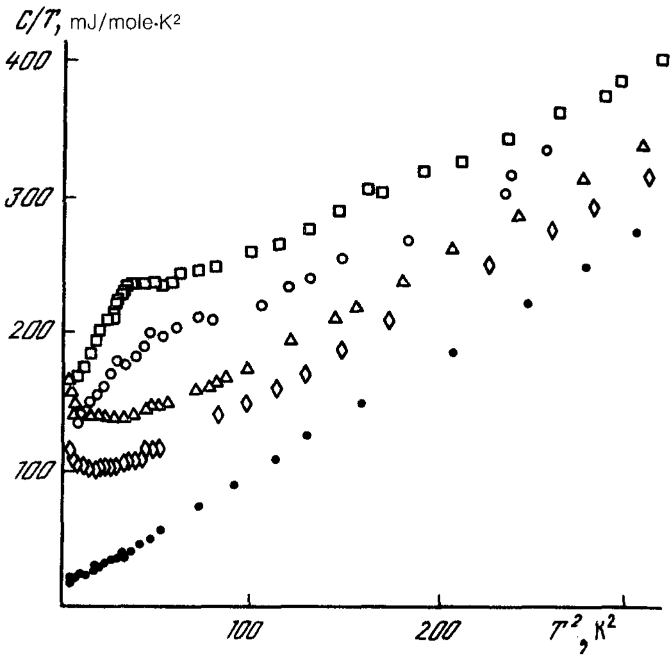


FIG. 2. Temperature dependence of the electronic component of the specific heat of $Ce_{1-x}La_xNiSn$ for the compositions with $x=0$ (□); $x=0.03$ (O); $x=0.1$ (△), $x=0.5$ (◇); and $x=1$ (●).

3. The temperature dependences of the magnetic contribution to the resistivity $\rho_m = \rho(\text{CeNiSn})-\rho(\text{LaNiSn})$ for the two CeNiSn samples and for the solid solutions $Ce_{1-x}La_xNiSn$ are plotted in Fig. 1. In the temperature interval $80 < T < 300$ K the $\rho_m$ $(T)$ curve for CeNiSn is characterized by a region in which the resistivity increases in a Kondo manner, $\rho \sim \ln T$. At temperatures in the range $T <$ 10–30 K a decrease in the temperature induces an exponential increase of the resistivity (see the inset in Fig. 1): $\rho_m \sim \exp(\epsilon_g)/2k_B T)$ with a parameter $\epsilon_g \approx$ (6–10) K. The temperature dependence of the electronic part of the specific heat of CeNiSn, $C/T = f(T^2)$, is shown in Fig. 2. Extrapolation of the "high-temperature" part of the $C/T$ curve to $T=0$ gives the value $\gamma \approx 200$ mJ/(mole·K$^2$), which is characteristic of the Kondo systems with $T_K \approx$ (30–50) K. At temperatures $T < 6$ K the $C/T$ curves exhibits a slight peak and $\gamma$ decreases as $T \to 0$. Replacement of some of the Ce atoms with La atoms leads to a suppression of the low-temperature dielectric state (Figs. 1 and 2).

The differential thermal emf of $S(T)$ and of CeNiSn, measured relative to copper, is characterized by a maximum near $T \approx 80$ K and by an increase in the values of $S$ in the region $T < 20$ K, where a gap develops near the Fermi energy. At high temperatures the Hall coefficient of CeNiSn is approximately equal to a value typical of normal metals and increases appreciably as the temperature is lowered in the region $T < 4.2$ K. At $T = 4.2$ K the Hall concentration of carriers is $n_H \approx 10^{21}$ cm$^{-3}$.

The application of hydrostatic pressure $p$ to 16.1 kbar initially induces a smooth

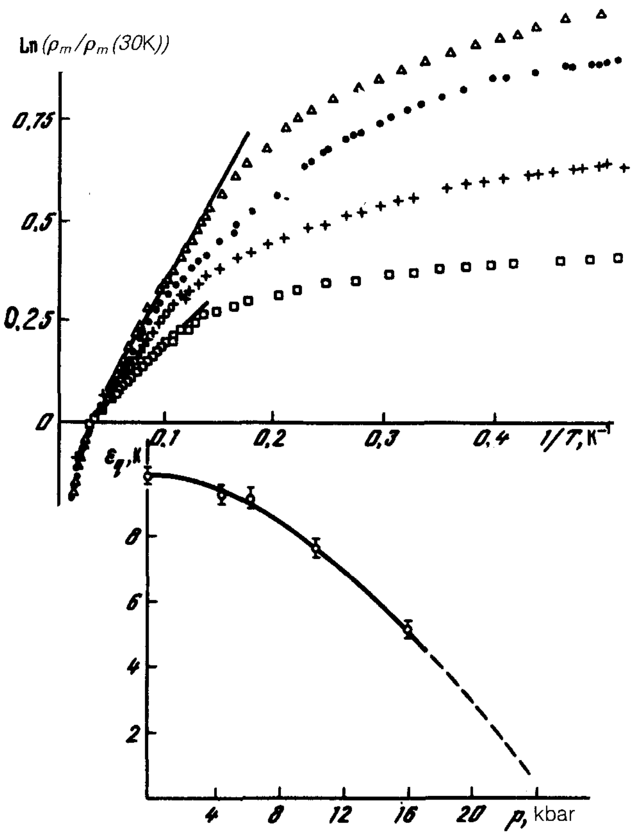


FIG. 3. Effect of hydrostatic pressure on the temperature dependence of the normalized component of the resistivity of CeNiSn (sample 58) in the coordinates of Ln$[\rho_M/\rho_M(30\text{ K})] = f(1/T)$ (△)—$p = 1$ bar; ●—6.5 kbar; +—10.5 kbar; □—16. kbar). The inset shows the pressure dependence of the energy gap, $\varepsilon_g(p)$.

decrease of $\epsilon_g$ (see the inset in Fig. 3) and then, at $p > 6$ kbar, a sharper increase at the rate $d\epsilon_g/dp \approx 0.45$ K/kbar. An extrapolation to $\epsilon_g = 0$ gives $p_k \approx 24$ kbar. After the pressure is removed, the original shape of the $\rho(T)$ curve is restored.

4. The appearance of a gap in the band spectrum of CeNiSn at $T < 10$ K cannot be the consequence of a magnetic transition, since the temperature dependence of the specific heat does not have the anomalies peculiar to this transition, while in the limit $T \to 0$ the magnetic susceptibility is characterized only by a slight deviation from the Curie-Weiss dependence $\chi(T)$. A conversion to a dielectric spectrum of CeNiSn at low temperatures may occur because in this compound the number of free electrons per magnetic Ce atom ($n$) is exactly equal to unity ($n = 1$). At $T \ll T_K$ the Fermi surface in this case should have zero volume, since the band should have two electrons and it should be filled completely if the $4f$ electrons are taken into account. Numerical modeling carried out in Refs. 11 and 12 has made it possible to show that satisfaction of the condition $n = 1$ induces the formation of a gap in $\epsilon_F$ and that the exponential

increases $\rho \sim \exp(\epsilon_g / 2k_B T)$ is virtually independent of the parameter of the *s-f* exchange coupling. Replacement of the magnetic Ce atoms with a nonmagnetic La atom in $Ce_{1-x}La_xNiSn$, which leads to a disruption of the $n = 1$ balance by increasing the value of $n(n > 1)$, induces a gradual suppression of the dielectric state and a transition to a logarithmic increase of $\rho(T)$ in the region of intermediate values of $x$. We note in conclusion that a transition of the spectrum to a dielectric state in the coherent regime has also been recently observed in $Ce(Pd_{1-x}Cu_x)_3$ with $x > 0.33$ (Ref. 13). In contrast with CeNiSn, however, disorder exists in this system because of the replacement of palladium with copper.


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