Anisotropic magnetic-field-induced crossover from a pseudogap to a heavy-fermion state in CeNiSn

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Using the break-junction tunneling technique in magnetic fields up to 20 T, a magnetic-field-induced crossover from a pseudogap to a metallic heavy-fermion state has been observed in CeNiSn. The pseudogap at the Fermi level which opens at $T<10$ K, was suppressed by a magnetic field applied along the a axis but was not affected for a field along the b axis. [S0163-1829(97)52512-4]

Compounds with rare-earth and actinide ions show strong electron correlations between the magnetic ions with interesting coherence effects due to the regular lattice positions of the magnetic ions. These so-called Kondo-lattice systems have a variety of ground states related to the hybridization between localized $4f$ electrons and conduction-band electrons, e.g., metallic heavy-fermion, semiconducting, superconducting, and/or magnetically ordered ground states.[1] They are characterized by their proximity to a magnetic instability, i.e., a transition where the system goes from a long-range magnetic order to a paramagnetic ground state as a function of a tuning variable like doping or pressure. In the case of CeNiSn, it was first believed that the ground state might be semiconducting as this compound has an even number of electrons. Experiments on better crystals lead now to the conclusion that CeNiSn is a metal with a finite density of states, which decreases sharply at the Fermi level E_F with a pseudogap behavior (here we call this system a Kondo pseudometal). The finite density of states at E_F yields a residual electronic specific heat $C=\gamma T$ with $\gamma=40$ mJ/K^2 mol as the temperature $T\rightarrow 0$ K whereas $\gamma=180$ mJ/K^2 mol at 8 K.[2]

A tunneling spectroscopy study of CeNiSn polycrystals gave direct evidence for a symmetric pseudogap structure of the broadened BCS type with a width $2\Delta=8-10$ meV.[3] We report here a direct observation of a very anisotropic suppression of the pseudogap in a break-junction tunneling experiment realized in a single crystal up to 20 T. The measurements in single crystals will give one the opportunity to clarify the strong anisotropic response to the magnetic field. The observed magnetic-field dependence of the density of states reflects the recovery of the heavy fermion character with a maximum in the density of states around E_F in high magnetic fields. The field dependence of the density of states is discussed in view of a metamagneticlike transition, which has been observed in several heavy-fermion compounds.

For our experiments we used two sets of samples (Nos. 1 and 2) prepared by different methods. The preparation of the No. 1 sample was done by annealing CeNiSn polycrystalline ingots at 1075 K for 2 weeks in purified argon to inhibit Sn evaporation and by subsequent annealing in UHV for few hours at the same temperature. The single-crystal domains were extracted from the annealed ingots using a spark cutter, and the crystal orientation was checked by Laue diffraction. Specific-heat measurements revealed no anomaly corresponding to impurity phases down to 80 mK. The estimated $\gamma=C/T(T=0\ \text{K})=35$ mJ/K^2 mol agrees with the result in single crystals purified by an electron transport method.[5] No increase of resistivity for the current $I\parallel c$ was observed at temperatures below 1 K and the residual resistivity ρ_a for $I\parallel a$ equals 80 $\mu\Omega$ cm. These facts give evidence for the high quality of the No. 1 sample. The No. 2 sample was prepared by the Czochralsky method. In these samples a small amount of parasitic phases has been detected in the specific-heat measurements. The γ and ρ_a values are, respectively, 50 mJ/K^2 mol and 150 $\mu\Omega$ cm.

Experiments were performed using the break-junction technique. The CeNiSn single crystals with In-soldered

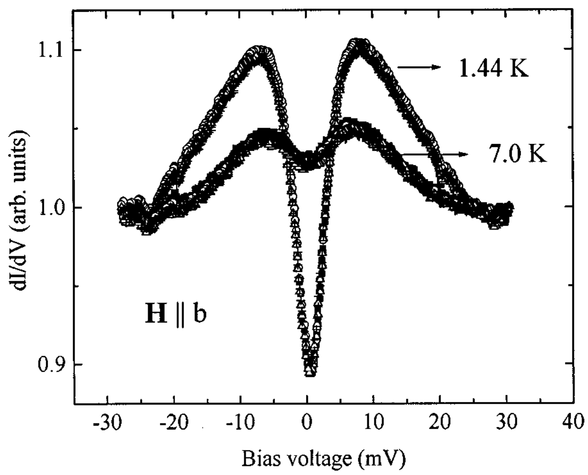

FIG. 1. $dI/dV(V)$ spectra of a CeNiSn (No. 1 sample) break junction at 0 (solid squares), 3 (open circles), 6 (open triangles), and 9.7 T (crosses) for 1.44 and 7.0 K with $H\|b$ and $I\|b$. Because of no field dependence, the symbols are difficult to distiguish.

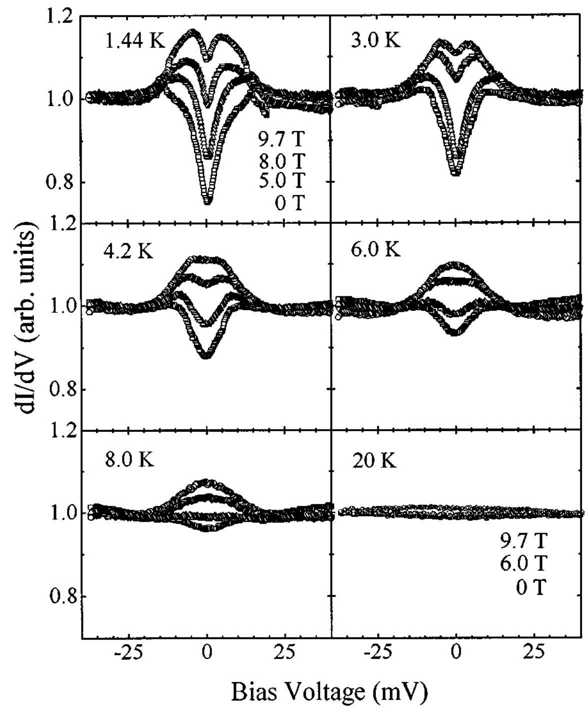

FIG. 2. $dI/dV(V)$ spectra of a CeNiSn (No. 1 sample) break junction at different magnetic fields from 0 to 9.7 T with $H\|a$ and $I\|b$ in the temperature range from 1.44 to 20 K.

electrical contacts were mounted on a flexible substrate and broken in liquid helium. Measurements were performed in the temperature range from 1.44 to 20 K and in magnetic fields up to 20 T. The junction conductance $dI/dV(V)$ has been measured using a standard lock-in modulation technique in the four-probe configuration. Measurements were performed on the junctions with resistance from 20 to 30 Ω. In this resistance range the junction resistance was very stable for variations of the magnetic field and the temperature. The observed gaplike structures were reproducible and always present for contact resistances $>10\ \Omega$.

The investigated range of junction resistances should normally correspond to a microcontact regime with metallic-type transport. However, as pointed out by Ekino *et al.*,[3] the observed features in the conductance curves can be interpreted as a measure for the electronic density of states as in a tunneling experiment. It should be noted that pseudogap structures have been observed at much larger tunnel-junction resistances $R>(2e^2/h)^{-1}$, i.e., the resistance for a direct single-atom contact. In addition, the junction zero-bias resistance is independent of the magnetic field applied parallel to the b axis where the magnetoresistance of bulk CeNiSn changes considerably in any field orientation ranging from 10 to 60%.[2,4] The tunneling regime may dominate even at the low contact resistance in the CeNiSn break junctions. Therefore we will analyze our results in terms of structure in the density of states. We note that the break junction's differential conductance is the convolution of the densities of states of the two similar electrodes.

In Fig. 1 we have shown the spectra at four different magnetic fields H from 0 to 9.7 T with $H\|b$ and $I\|b$ for the No. 1 sample (since the current direction cannot be properly defined locally in the contact area, we indicate with this notation the current direction before breaking). A gap structure symmetric with respect to bias polarity has been observed in the voltage dependence of the conductance $dI/dV(V)$ of the CeNiSn break junction. The gap begins to open at temperatures below $T\simeq 10$ K and its depth and peak-to-peak separation increase as the temperature is lowered. By analogy with superconductor-insulator-superconductor junctions, the peak-to-peak separation in the break-junction geometry defines the 4Δ value yielding a gap width $2\Delta=8-10.5$ meV. Our results at zero magnetic field are in a good agreement with recent break-junction measurements of CeNiSn polycrystals.[3] In contrast to the break-junction study of polycrystalline CeNiSn where both 2Δ and 4Δ values for the peak-to-peak separation were observed, we always observed only the 4Δ peak-to-peak separation for the CeNiSn single crystals. It is remarkable that no influence of the magnetic field is observed up to 10 T in the whole temperature range from 1.44 to 20 K, indicating that the density of states is independent of magnetic field along the b axis. This result is consistent with specific-heat measurements.[2]

On the contrary, for $H\|a$, we see a strong magnetic field dependence as shown in the data of Fig. 2 for the No. 1 sample ($I\|b$). The gap is suppressed by a magnetic field and transforms into a heavy-mass resonance peak around zero bias voltage, which is clearly developed at higher temperatures. For lower temperatures the 10-T magnetic field is not sufficient to suppress the pseudogap completely. In the inset of Fig. 3 we show the result for the No. 2 sample at 1.7 K for higher fields $H\|a$ up to 20 T. The split-peak structure vanishes completely at a magnetic field around 14 T.

Figure 3 shows the field dependence of the square root of the zero-bias conductance $[dI/dV(0)]^{1/2}$ for the No. 2 sample which corresponds to the density of states at E_F. In a break-junction tunneling experiment the current can be written as an energy convolution of two density of states functions with energy difference eV. For smooth functions around zero bias voltage, $[dI/dV(0)]^{1/2}$ equals the density of states at the Fermi level. For comparison, we have shown the specific-heat data C/T for CeNiSn obtained at 0.76 K.[2]

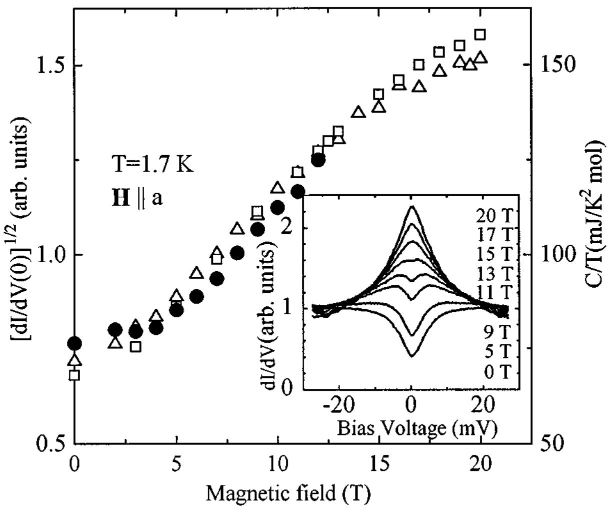

FIG. 3. Square root of the zero-bias conductance $dI/dV(0)$ as a function of magnetic field $\|a$ for break junctions of two different pieces of the No. 2 sample (triangle and square symbols). The solid circles show the specific-heat data (right axis) taken from Ref. 2 The inset shows the $dI/dV(V)$ curves for fields $\|a$.

Both the magnetic-field dependence of $[dI/dV(0)]^{1/2}$ and its relative variation are in agreement with the specific-heat studies and clearly indicate the transition from the Kondo pseudometal to the metallic heavy-fermion state when the external magnetic field is applied along the a axis.

The observed suppression of the gap by a magnetic field parallel to the a axis is consistent with a previously proposed model.[6] In this model, the magnetic-field-induced increase of the density of states is explained in terms of the Zeeman spin splitting of the quasiparticle band at the Fermi level. Because the magnetic susceptibility is highly anisotropic ($\chi_a \gg \chi_b, \chi_c$) in this compound,[5] the induced spin polarization occurs only when the magnetic field is applied along the a axis. Thus the density of states is expected to increase only in the case of $H\|a$ by this model. This Zeeman-splitting model qualitatively explains the present results. However, the model predicts that the density of states decreases with increasing magnetic field at high temperatures[6] ($T>7$ K) whereas our results show an increase of $dI/dV(0)$ with magnetic field over the whole investigated temperature range (see Fig. 4).

Since the magnetic field usually modifies the antiferromagnetic intersite (RKKY) interaction in Kondo lattices, the dispersion of the quasiparticle band (or, equivalently, mass enhancement) depends on magnetic field as observed in other heavy-fermion compounds (e.g., $CeRu_2Si_2$).[7] This picture can give another way the crossover from the pseudometallic to the heavy-fermion state by a magnetic field can occur and can also explain our results qualitatively.[8] The modification of the dispersion is effective when $\mu_B H \simeq k_B T^*$, where T^* is the characteristic temperature of the coherent state at low temperatures.[8] For the estimated $T^* \simeq 10$ K (see below), this condition can be fulfilled for the present field study up to 20 T. Probably both the bare Zeeman splitting and the modification of the dispersion, i.e., of the coherent effect, have to be included in a microscopic model in order to interpret the observed anisotropic effect of the magnetic field quantitatively.

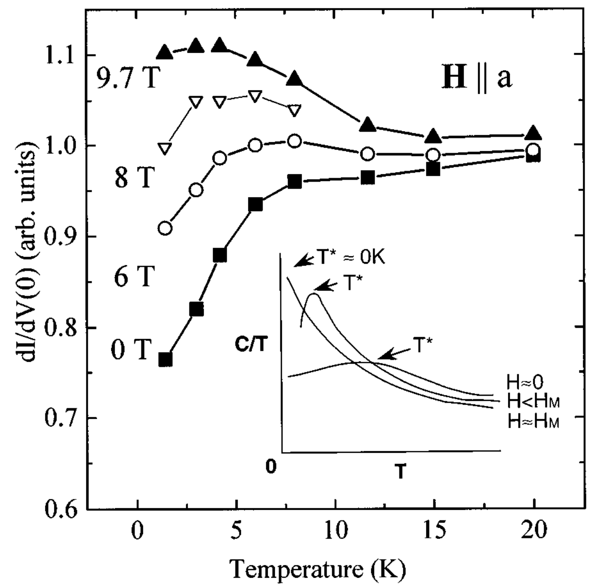

FIG. 4. Zero-bias conductance $dI/dV(0)$ behavior of a CeNiSn (No. 1 sample) break junction as a function of temperature for different magnetic fields $\|a$. For comparison, the inset shows in a schematic way the field dependence of $C/T(T)$ for $H<H_M$ in a Kondo-lattice system like $CeRu_2Si_2$.

Usually, in metallic heavy-fermion systems, the density of states increases with magnetic field and starts to decrease at a critical field H_M (metamagneticlike transition) where the Zeeman energy $2\mu_B H_M$ overcomes the antiferromagnetic correlation energy Δ_{AF}. For example, in $CeRu_2Si_2$, the association of $H_M=7.8$ T with the collapse of the antiferromagnetic correlation has been confirmed directly by the neutron scattering measurements; the Zeeman energy $2\mu_B H_M$ roughly corresponds to $\Delta_{AF} \simeq 1$ meV.[11] In $CeRu_2Si_2$, C/T (a measure for the density of states) reveals a magnetic-field dependence in the temperature dependence as shown schematically in the inset of Fig. 4.[9,10] The temperature T^* at the maximum in C/T decreases from $\simeq 5$ K at zero field to approximately zero for fields up to H_M. Above H_M, T^* increases again.

Figure 4 represents the zero-bias conductance $dI/dV(0)$ as a function of temperature for different magnetic fields. At $H=0$ T, the decrease of the zero-bias conductance for $T<7$ K is due to the pseudogap formation. T^* defined by the maximum in the T dependence of $dI/dV(0)$ shows the precursor behavior for a metamagnetic transition decreasing from $\simeq 10$ K at 0 T, to $\simeq 8$ K at 6 T, and then to $\simeq 3$ K at 9.7 T (see Fig. 4). With the continuous increase of $dV/dI(0)$ up to 20 T shown in Fig. 3, the metamagnetic transition is not observed in CeNiSn, i.e., $H_M>20$ T. As in $CeRu_2Si_2$, the collapse of the antiferromagnetic correlation by a magnetic field can be of importance: Δ_{AF} in CeNiSn appears to be $\simeq 4$ meV as obtained from inelastic neutron scattering[12] yielding $H_M=\Delta_{AF}/2\mu_B \simeq 30$ T. This value is supported by the saturation of $dI/dV(0)$ around 20 T (Fig. 3).

A particular feature in CeNiSn is that the large gap ($2\Delta/k_B \simeq 80$ K) opens at much lower temperatures ($\simeq 10$ K) and closes significantly by a small magnetic field (14 T corresponds to 10 K). Such a property may be interpreted by analogy with strong-coupling superconductivity where the gap energy is considerably larger than the value $3.5k_BT_c$ for the weak-coupling case (T_c the superconducting transition temperature). We emphasize here that $T^*\simeq 10$ K and $\Delta_{AF}\simeq 4$ meV govern the scaling for the magnetic-field and temperature dependence of the gap in CeNiSn. We expect that the field dependence of the density of states in the Kondo lattices, such as CeNiSn, $CeRu_2Si_2$, and $CeCu_6$, can be explained by a unified picture characterized by k_BT^* and Δ_{AF} for the energies of, respectively, the coherent electron system and the antiferromagnetic correlations. Even one energy scale, e.g., Δ_{AF}, may be sufficient since a linear relation between $H_M\simeq\Delta_{AF}/2\mu_B$ and T^* at zero field has been observed in a series of doped $Ce_{1-x}La_xRu_2Si_2$.[9]

Investigations of the physical meaning of the $2\Delta = 8-10$ meV excitations and their relation with the spin excitations, i.e., with Δ_{AF}, in CeNiSn are to be focused. Kagan, Kikoin, and Mishchenko proposed that in CeNiSn a pseudogap opens in the spin excitation spectrum and explained the neutron scattering spectra quantitatively.[13] However, the charge excitation spectrum and its field dependence have not been calculated based on their theory.

Let us finally compare data with the response to the magnetic field in mixed valence compounds like TmSe and YbB_{12}, which are well known to have an insulating ground state. In TmSe, the observed tunneling gap ($2\Delta = 1.2$ meV) appears only when the antiferromagnetism sets in below $T_N = 3.5$ K and disappears completely in a magnetic field $H_c = 1.3$ T.[14] Because 2Δ, H_c, and T_N are of the same order, this behavior can be categorized as a weak-coupling property due to a simple exchange splitting of the rigid band by the ordered moment and its collapse by the Zeeman effect. The same rigid band picture is also supported by high-field magnetoresistance measurements in nonmagnetic YbB_{12}.[15] It will be worthwhile in the future to observe how the tunneling gap depends on microscopic parameters such as the Kondo temperature and the antiferromagnetic correlation energy Δ_{AF}.

In conclusion, the tunneling spectroscopy was applied to study the density of states around the Fermi level in CeNiSn. A symmetric pseudogap with respect to the Fermi level has been always observed at $T<10$ K and found to be strongly suppressed by a magnetic field if only applied parallel to the *a* axis. Above the crossover field the gap vanishes and an enhancement of the density of states at the Fermi level is observed, indicating the transition to a metallic heavy-fermion state.

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