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Uniaxial-stress induced magnetic order in CeNiSn

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We report the observation of an unexpected magnetic transition in the Kondo semimetal CeNiSn under uniaxial pressures. For $P\|a$, both the specific heat C and magnetic susceptibility χ decrease steadily with increasing P. By contrast, once $P\|b$ and $P\|c$ exceed 0.13 GPa, $C(T)$ exhibits a distinct jump at 4 K followed by a peak at 3 K. The concomitant peaking in $\chi(T)$ for $P\|B\|c$ is indicative of an antiferromagnetic transition. [S0163-1829(99)51234-4]

Pressure can turn a magnetically ordered state to a nonordering state in strongly correlated electron systems such as heavy-fermion compounds and transition-metal oxides by tuning the electronic couplings and band width.[1] For example, in a cerium-based antiferromagnet at ambient pressure, the intersite Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction dominates the on-site Kondo interaction.[2] Under pressure, however, the Kondo interaction is enhanced more strongly than the RKKY interaction, leading to a magnetic instability. The approach from a nonmagnetic state to the magnetic instability under pressure has been realized only in certain ytterbium-based compounds.[3] Recently, much interest has focused on anomalous phenomena at the critical pressure, i.e., non-Fermi-liquid behavior[4] and unconventional superconductivity.[5]

Most Ce-based compounds being away from the critical point possess a metallic ground state. However, CeNiSn forms a pseudogap in the density of states at the Fermi level.[6] The magnitude of the energy gap was derived from nuclear magnetic resonance and specific-heat measurements to be 14 and 21 K, respectively.[7,8] The anisotropic behavior in magnetic and transport properties originates from the strong c-f hybridization of the Ce $4f$ states with the Ni $3d$ and Sn $5p$ band states in the orthorhombic structure.[9] The magnetic susceptibility along the a axis, χ_a, is much larger than χ_b and χ_c, and exhibits a pronounced peak at 12 K.[6] Below this temperature, dynamic antiferromagnetic correlations develop,[10] but no transition into static magnetic order occurs even at 10 mK.[11]

A systematic study has shown that the gap formation in CeNiSn is very sensitive to the degree of c-f hybridization.[9,12–15] Partial substitution of Cu, Pd, and Pt for Ni strongly suppresses the pseudogap,[9] leading to a long-range magnetic order at low temperatures for 8 at. % Cu and 33 at. % Pt.[12,13] These substitutions expand the unit cell volume, which may weaken the c-f hybridization. Under a hydrostatic pressure, by contrast, the c-f hybridization should be strengthened. However, the pseudogap is also suppressed as was indicated from the decrease in the absolute value of the Hall coefficient.[14] This pressure effect suggests that the carrier concentration increases together with the recovery of the density of states at the Fermi level. Furthermore, suppression of antiferromagnetic correlations upon applying pressure was deduced from a neutron inelastic-scattering experiment.[15] It should be recalled that noncubic heavy-fermion compounds such as $CeCu_{5.8}Au_{0.2}$ (Ref. 16) and UPt_3 (Ref. 17) exhibit significantly different responses to an uniaxial pressure from that to a hydrostatic pressure. Therefore, one expects an anisotropic response of the pseudogap in CeNiSn to a uniaxial pressure. In this paper, we report the observation of a magnetic ordering in CeNiSn under uniaxial pressures by the measurements of the specific heat and magnetic susceptibility.

The single crystal of CeNiSn was prepared by a Czochralski method using a hot tungsten crucible in a radio-frequency induction furnace. Details of the preparation methods and characterization of the crystals were described elsewhere.[18] The specific heat under uniaxial pressures up to 0.4 GPa was measured using an ac method in the temperature range $1.7 \leqslant T \leqslant 20$ K. A disk-shaped $3.0\,\mathrm{mm}\phi \times 0.2$ mm sample was cut perpendicular to the principal axis. The sample was sandwiched between two Cu plates, on which a thermometer and a heater were mounted, respectively. A pair of pistons was made of ZrO_2 with a rather low thermal conductivity. To achieve better thermal isolation, diamond powder was placed between the Cu plate and the piston. The pressure was determined by the known pressure dependence of the superconducting transition temperature $T_c(P)$ of indium. The transition was measured by the mutual induction method with coils outside the pressure cell. The contribution of the Cu plates, thermometer, and heater to the total heat capacity was determined in separate measurements without the sample. We found that this contribution is 10–20 % of the total heat capacity for $2 \leqslant T \leqslant 10$ K, and is essentially independent of pressure up to 0.3 GPa. The absolute value of the specific heat of the sample was yielded by comparing with the value obtained by an adiabatic method at ambient pressure.[10] The magnetic susceptibility under uniaxial pressure for $B\|P$ was measured by a superconducting quantum interference device magnetometer in the range $2 \leqslant T \leqslant 10$ K. The pressure cell and pistons were, respectively, made of hardened Cu-Be alloy and ZrO_2. The pressure was determined by the known dependence of $T_c(P)$ for tin. Details of the experimental setup are described in Ref. 19.

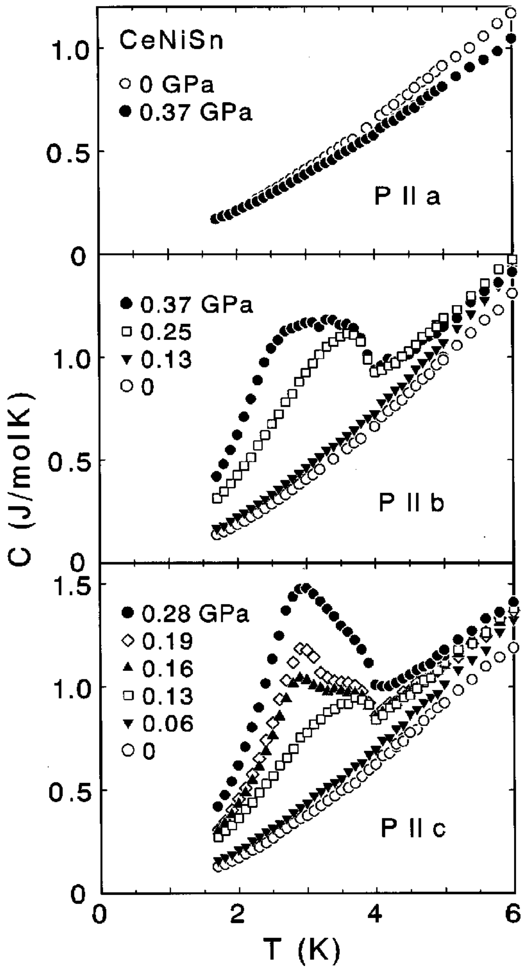

FIG. 1. Specific heat C of CeNiSn as a function of T under uniaxial pressures. Here, a clear sign of a phase transition is visible for $P\|b \geqq 0.25$ GPa and $P\|c \geqq 0.13$ GPa.

Figure 1 shows the temperature dependence of the specific heat $C(T)$ of CeNiSn for several pressures parallel to the *a*, *b*, and *c* axes. For $P\|a$, the value of C decreases slightly with P in the whole temperature range. By contrast, for $P\|b$ and $P\|c$, $C(T)$ increases with P and exhibits a maximum around 3.7 K at 0.25 GPa and 0.13 GPa, respectively. With a further increase in P, another peak develops around 3 K. We note that the double-peak structure is not due to a gradient in the pressure field because a number of measurements on many samples with different dimensions reproduced the data shown in Fig. 1. A zero-pressure recheck for samples pressurized above 0.2 GPa showed no evidence for irreversible effect or the presence of residual strains.

The magnetic susceptibility χ at various uniaxial pressures is shown in Fig. 2. For $P\|B\|a$, the value of χ decreases with P, whereas the maximal temperature at 13 K hardly changes. By contrast, for $P\|B\|c$, χ is largely enhanced with development of a pronounced maximum at 4 K above 0.25 GPa. The concomitant peaking in both $C(T)$ and $\chi(T)$ clearly indicates that an antiferromagnetic (AF) order sets in. This is consistent with the T^3 dependence of $C(T)$ below 2.5 K for $P\|c$, which can be regarded as characteristic of the excitation of AF magnons.

We turn now to examine how the pseudogapped state is replaced by the magnetically ordered state. We estimated the

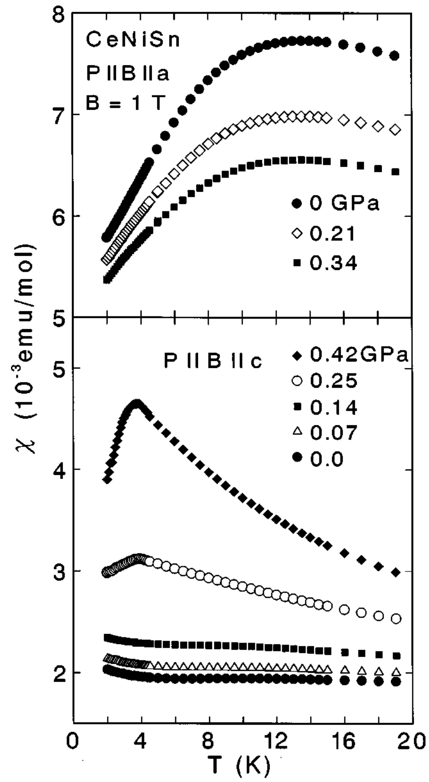

FIG. 2. Magnetic susceptibility χ of CeNiSn as a function of temperature for different uniaxial pressures $P\|B\|a$ and $P\|B\|c$. The peak at 4 K for $P\|c \geqq 0.25$ GPa indicates an antiferromagnetic transition.

electronic contribution to the specific heat C_{el} by subtracting the phonon part for LaNiSn (Ref. 8) from the measured value for CeNiSn. The data of C_{el}/T for selected pressures are shown in Fig. 3. For $P\|c = 0.16$ GPa, the coexistence of the

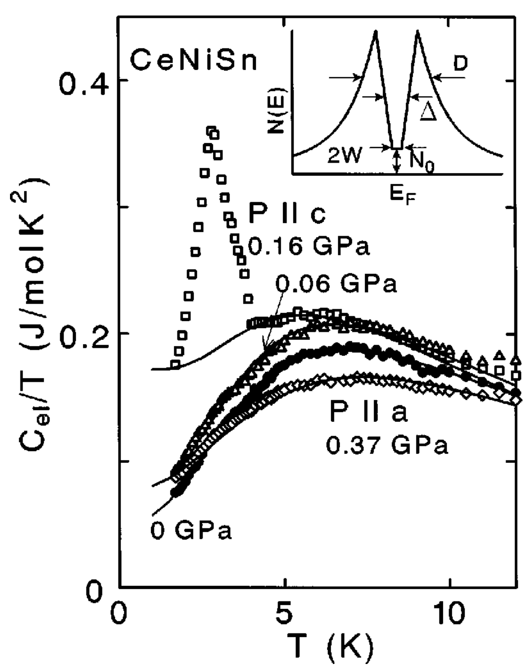

FIG. 3. Electronic specific heat divided by temperature C_{el}/T of CeNiSn for $P\|c = 0$, 0.06, and 0.16 GPa and for $P\|a = 0.37$ GPa. Solid lines are the fits with a V-shaped density of states depicted in the inset.

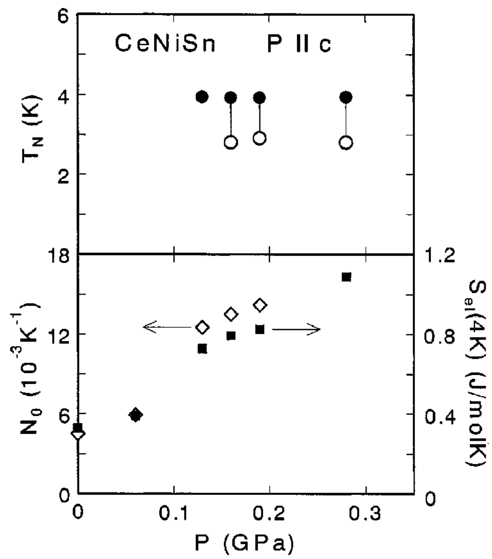

FIG. 4. The magnetic transition temperature T_N, the residual density of states N_0 (◇), and the electronic entropy $S_{\rm el}$ at 4 K (■) as a function of uniaxial pressure for $P\|c$. In the upper panel, the closed and open circles stand for, respectively, the temperatures at the jump and at the peak in the data of $C(T)$ plotted in Fig. 1.

maximum around 6 K with the peak at 3 K implies that the AF state evolves from the state where the pseudogap remains. As is shown by the solid curves, the temperature variations of $C_{\rm el}/T$ in the nonordered state could be reproduced by the use of the V-shaped density of states (DOS) with a finite value at the Fermi level.[7,8] The DOS is illustrated in the inset of Fig. 3, where the parameters D, Δ, and W are the half-widths of the main DOS, the V-shaped gap, and the bottom of the gap, respectively, and N_0 is the residual DOS at the Fermi level. If we assume the condition $\int N(E)dE=1$ to be held at all pressures, then it is found that both D and Δ increase with $P\|a$ while W and N_0 hardly change up to 0.37 GPa. With thus obtained parameters, we could reproduce the pressure dependence of $\chi_a(T)$, as was done at ambient pressure.[20] This analysis showed that the observed depression of $\chi_a(T)$ with $P\|a$ is a result of the increase of D, i.e., increase in the c-f hybridization.

We show in Fig. 4 the dependence of the AF transition temperature T_N on the applied pressure for $P\|c$. Because of the double-peak structure in $C(T)$, we plot here the two temperatures at the initial jump and at the maximum in $C(T)$. The most significant feature is that once T_N appears at 4 K above 0.1 GPa, it does not change with further increase in P. This fact indicates the pressure-induced transition to be a first-order transition. In the lower panel of Fig. 4, we compare the P dependence of N_0 with that of the electronic entropy $S_{\rm el}$ at 4 K. We estimated $S_{\rm el}(T)$ below 1.7 K in the magnetically ordered state on the assumption of $C_{\rm el}(T)=\gamma T+\alpha T^3$. From Fig. 4, it is clear that the nonmagnetic-magnetic transition at 0.1 GPa is well correlated with the sudden increase in both N_0 and $S_{\rm el}(T=4\,{\rm K})$. This fact suggests that the increase in N_0 with P triggers the magnetic transition.

We have shown, so far, that CeNiSn undergoes a transition from a pseudogapped state to a magnetically ordered state under uniaxial pressure for $P\|b$ and $P\|c$, while no transition occurs under $P\|a$. Since the critical pressure for this transition is as small as 0.1 GPa, CeNiSn should be located in the vicinity of the quantum critical point. As to the microscopic origin of the anisotropic response, we recall that the local symmetry of Ce ions in this compound is quasitrigonal when viewed from the orthorhombic a direction.[21] This trigonal symmetry is thought to be a prerequisite for formation of the pseudogap in the renormalized band which is originated from the anisotropic c-f hybridization.[22,23] When a uniaxial pressure is applied along the b and c axes, the trigonal symmetry would be lowered, while it is maintained for $P\|a$. The loss of the quasitrigonal symmetry may be responsible for the pressure-induced magnetic transition. Another scenario is that the elongation along the a axis under $P\|b$ and $P\|c$ may weaken the c-f hybridization and thus brings the system to the magnetically ordered state, as is expected from Doniach's phase diagram.[2] To check this scenario, we measured the change in lattice parameters under uniaxial pressure at room temperature by using a strain-gauge technique. It is found that the a parameter actually increases for $P\|c$ at a rate of $(1/a_0)da/dP=0.013\,{\rm GPa}^{-1}$, whereas it decreases for $P\|b$ at a rate of 0.0024 GPa^{-1}. However, even at ambient pressure, anisotropic variations of the lattice parameters take place with decreasing temperature.[24] Therefore, a crystallographic study at low temperatures under uniaxial pressures remains to be performed to check the possible structural transition. In any event, pressure-induced magnetism has been observed in CeNiSn.

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[1] See, e.g., J. D. Thomson and J. M. Lawrence, in *Handbook on the Physics and Chemistry of Rare Earths*, edited by K. A. Gschneidner, Jr. *et al.* (North-Holland, Amsterdam, 1994), Vol. 19, p. 383.

[2] S. Doniach, Physica B & C **91B**, 231 (1977); J. R. Iglesias, C. Lacroix, and B. Coqblin, Phys. Rev. B **56**, 11 820 (1997).

[3] K. Alami-Yadri, H. Wilhelm, and D. Jaccard, Eur. Phys. J. B **6**, 5 (1998).

[4] H. v. Löhneysen *et al.*, Phys. Rev. Lett. **72**, 3262 (1994); K. Umeo, H. Kadomatsu, and T. Takabatake, Phys. Rev. B **55**, R692 (1997).

[5] D. Jaccard *et al.*, Rev. High Pressure Sci. Technol. **7**, 412 (1998); E. Vargoz *et al.*, Physica B **229**, 225 (1997); N. D. Mathur *et al.*, Nature (London) **394**, 39 (1998).

[6] T. Takabatake *et al.*, Phys. Rev. B **41**, 9607 (1990).

[7] M. Kyogaku *et al.*, J. Phys. Soc. Jpn. **59**, 1728 (1990).

[8] S. Nishigori *et al.*, J. Phys. Soc. Jpn. **65**, 2614 (1996).

[9] T. Takabatake *et al.*, J. Magn. Magn. Mater. **177-181**, 277 (1998).

[10] T. E. Mason *et al.*, Phys. Rev. Lett. **69**, 490 (1992); H. Kadowaki *et al.*, J. Phys. Soc. Jpn. **63**, 2074 (1994).
[11] A. Kratzer *et al.*, Europhys. Lett. **19**, 649 (1992).
[12] A. Brückl *et al.*, Physica B **240**, 199 (1997).
[13] S. Nishigori *et al.*, Physica B **186-188**, 406 (1993).
[14] T. Hiraoka *et al.*, Physica B **199 & 200**, 440 (1994).
[15] T. J. Sato *et al.*, J. Phys.: Condens. Matter **8**, 8183 (1996).
[16] M. Sieck, F. Huster, and H. v. Löhneysen, Physica B **230-232**, 583 (1997).
[17] D. S. Jin *et al.*, Phys. Rev. Lett. **68**, 1597 (1992).
[18] G. Nakamoto *et al.*, J. Phys. Soc. Jpn. **64**, 4834 (1995).
[19] Y. Uwatoko *et al.*, Rev. High Pressure Sci. Technol. (to be published).
[20] K. Izawa *et al.*, J. Phys. Soc. Jpn. **65**, 3119 (1996).
[21] I. Higashi *et al.*, J. Alloys Compd. **193**, 300 (1993).
[22] H. Ikeda and K. Miyake, J. Phys. Soc. Jpn. **65**, 1769 (1996).
[23] Yu. Kagan, K. A. Kikoin, and A. S. Mishchenko, Phys. Rev. B **55**, 12 348 (1997).
[24] Y. Uwatoko *et al.*, J. Magn. Magn. Mater. **104-107**, 643 (1992).