Effective carrier doping by magnetic field into a pseudogapped state in CeNiSn: A Sn NMR study

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(Received 9 April 1996)

The magnetic field dependence of the spin-lattice relaxation rate $1/T_1$ of ^{119}Sn in CeNiSn has been measured down to 20 mK in a field range of 0.2–8 T. $(T_1T)^{-1}$ is constant below 1 K, which depends on the external field in such a manner that its value stays constant to 2 T, while it increases linearly with the field up to 8 T. It is shown that this magnetic field dependence of $(T_1T)^{-1}$ is well explained by the simple scenario that the quasiparticle density of states at the Fermi level is produced by the Zeeman splitting of the up- and down-spin bands, keeping its V-shaped gapped structure unchanged for fields less than 8 T. The present experiment has elucidated that CeNiSn is in a semimetallic ground state with a low carrier density and the application of a magnetic field exceeding 2 T turns out to supply effective carriers. [S0163-1829(96)09933-X]

Ce-based compounds such as CeNiSn, CeRhSb, and $Ce_3Bi_4Pt_3$ have been reported to possess a novel energy gap. The gap of $Ce_3Bi_4Pt_3$ is of the activated type with the same order of the magnitude ($\sim$70 K) as in the compounds reported so far,[1] whereas it was argued from various measurements of the magnetic susceptibility, the resistivity, and the thermopower on single-crystal, CeNiSn and CeRhSb that a small gap opened in the quasiparticle band with the magnitude of several kelvin.[2–4] It has been controversial whether or not the ground state of CeNiSn is in a semiconducting state affected by impurities or a semimetallic state with very low carrier density accompanied by density of states (DOS) with a V-shaped pseudogap. From previous systematic NMR experiments on the single-crystal CeNiSn and substituted systems, it has been demonstrated that the tiny DOS is present inherently at the Fermi level located at the bottom in the V-shaped gapped structure as illustrated in Fig. 1(a), and that CeNiSn is classified as a semimetal with a very low carrier density.[5] It was found that the respective substitution of La and Co for Ce and Ni sites generated the DOS plateau over a finite energy range *near* the Fermi level. Interestingly, depending on whether holes or electrons are doped into the renormalized quasiparticle bands, the semimetallic state changes into either the normal Fermi liquid state or the antiferromagnetic ground state.[5]

In addition to the carrier doping effect, a further experiment has been currently planned in order to investigate to what extent the V-shaped pseudogap in CeNiSn is modified by the application of a magnetic field stronger than $\sim$1.5 T below which any field dependence of physical quantities was not reported thus far. An NMR experiment at high frequencies amounting to $\sim$130 MHz at 8 T has, however, not been established yet at very low temperatures down to 20 mK, which is a necessary condition to make clear an intimate change of the structure of the DOS curve. Actually, to our knowledge, this was a first trial in solid-state NMR so far. Nevertheless, it has been done successfully as presented in this paper.

From the magnetization measurements, it was reported that the gapped state in YbB_{12} was completely suppressed by

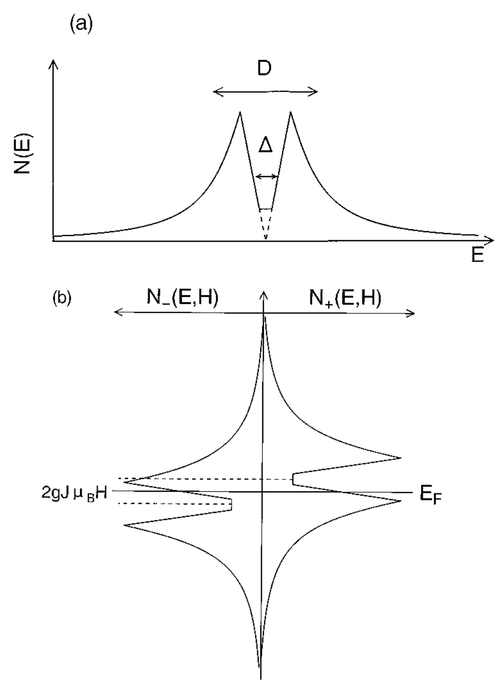

FIG. 1. (a) Schematic density of states (DOS) with the V-shaped pseudogap. Bandwidth, D=140 K, magnitude of gap, Δ=14 K, and N_{res}/N_0=0.077 were obtained in the previous work (Ref. 5). (b) Schematic DOS split by magnetic field. $N_+(E,H)$ and $N_-(E,H)$ are the DOS of up- and down-spin bands, respectively.

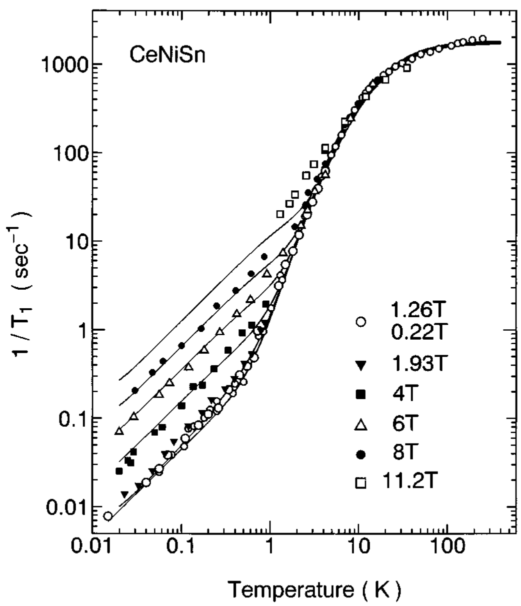

FIG. 2. Magnetic field dependence of $1/T_1$ of ^{119}Sn in CeNiSn. The temperature dependence of $1/T_1$ at the field of 0.22 and 1.26, 1.93, 4, 6, 8, and 11.2 T is indicated by open circles, solid squares, open triangles, solid circles, and open squares, respectively.

a high magnetic field as large as 50 T. Similarly the squeezing of the gap was observed in SmB_6 as well, although the influence is smaller than that in YbB_{12} because of the smaller g value of SmB_6.[6,7] The closing of the gap was argued to be due to the overlapping of each valence and conduction band split by the Zeeman shift. In CeNiSn with a small pseudogap, a significant magnetic field dependence of various physical quantities is anticipated in a relatively low field. Actually, the T-linear coefficient of the specific heat, γ_{res}, related to the DOS was nearly independent of the field up to 4 T, whereas it increased appreciably with further increasing the field.[8] A recent magnetoresistance experiment seems to be consistent with the picture that CeNiSn is semimetallic with a very low carrier density and the DOS increases by a magnetic field over 2 T.[9]

In this paper, in order to elucidate the magnetic field dependence of the V-shaped pseudogapped state in CeNiSn, we report the magnetic field dependence of $1/T_1$ of ^{119}Sn. The same single-crystal as in the previous works was employed in this work.[5] For NMR measurements, the single-crystal CeNiSn in Ref. 4 was crushed into small grains with diameter less than 70 μm to avoid the skin-depth effect of radio frequency (rf) fields. Then each grain was aligned so that the a axis is parallel to the magnetic field. The nuclear spin-lattice relaxation time T_1 of ^{119}Sn in CeNiSn was measured by observing the spin-echo recovery after a saturation pulse with the use of a homemade phase-coherent pulsed-NMR spectrometer. T_1 measurements in magnetic fields of 0.22, 1.26, 1.93, 4, 6, and 8 T were performed down to 20 mK using a ^{3}He-^{4}He dilution refrigerator. The sample was directly immersed into the ^{3}He-^{4}He mixture to avoid heating up due to the eddy current caused by the excitation rf pulse.

Figure 2 indicates the temperature dependence of $1/T_1$ of ^{119}Sn in CeNiSn under various magnetic fields (frequencies) of 0.22 T (3.5 MHz), 1.26 T (20 MHz), 1.93 T (31.4 MHz), 4 T (64.9 MHz), 6 T (98.1 MHz), and 8 T (132.6 MHz) down to 20 mK together with the data at 11.2 T (182 MHz) down to 1.3 K. $1/T_1$ at 0.22 and 1.26 T starts to decrease largely below 30 K and obeys a T_1T=const relation from 0.4 K to 20 mK, whereas with increasing magnetic field to 4, 6, and 8 T, the T_1T=const relation becomes apparent below 1, 2, and 3 K, respectively, although $1/T_1$ above $\sim$ 8 K is nearly independent of the magnetic field. It is substantial that the magnitude of $(T_1T)^{-1}$ is almost invariant up to 2 T and starts to increase progressively with further increasing field. A significant enhancement of $(T_1T)^{-1}$ reveals that the DOS is generated at the Fermi level by applying a magnetic field over 2 T. It is noteworthy that the DOS increased by the substitution of La for the Ce site and of Co and Cu for the Ni site, i.e., by doping either holes and electrons.[5] We note that $1/T_1$ at 11.2 T is somewhat reduced in the higher-T region than 10 K, whereas below 10 K it is markedly enhanced, exhibiting a rather different behavior from those in lower fields than 8 T.

In order to account for the field dependence of $1/T_1$, we take a simple model that the V-shaped quasiparticle band is split into up- and down-spin bands with a relative shift of $\Delta(k_B)=2gJ\mu_BH$ by the Zeeman interaction as illustrated in Fig. 1(b). By applying the magnetic field, the Fermi level crosses the E-linear line for each up- and down-spin band over the DOS plateau, and eventually, the DOS increases at the Fermi level. Since the energy over which the DOS has the plateau corresponds to the magnetic field of $\sim$ 2 T, the DOS is predicted to increase when exceeding 2 T. This simple picture is actually consistent with the enhancement of $1/T_1$ by a magnetic field over 2 T as seen in Fig. 2. In order to embody this, we have calculated the overall temperature dependence of $1/T_1$ following the relation of

$$\frac{1}{T_1}\propto A_{\mathrm{hf}}^2 T\int N_+(E-gJ\mu_BH)N_-(E+gJ\mu_BH)\times\left\{-\frac{\partial f(E)}{\partial E}\right\}dE. \tag{1}$$

The solid lines in Fig. 2 are calculations with the same parameters of D=140 K, Δ=14 K, a fraction of the residual DOS, $N_{\mathrm{eff}}(E_F)/N_0(E_F)$=0.077 as in the previous works in Refs. 10 and 11, and a parameter gJ=0.77 for the interpretation of the field dependence. It is remarkable that all the data for 0.22, 1.26, 1.93, 4, 6, and 8 T are in excellent agreement with the calculation. The above simple picture was, however, not applied to $1/T_1$ data at 11.2 T, suggesting that the pseudogap structure is no longer maintained in a stronger magnetic field than 10 T. We note that gJ is consistent neither with gJ=1.29 expected from the dominant $|5/2,\pm3/2\rangle$ state near the Fermi level, which was pointed out theoretically,[12] nor with gS=1 for free electrons. This result suggests that the quasiparticle state is hybridized not only with $|5/2,\pm3/2\rangle$, but also mixed with other states.

Next, to extract an increasing rate of the DOS at the Fermi level, E_F, by applying a magnetic field, we note the relation

$$\sqrt{\frac{[T_1T(H)]^{-1}}{(T_1T)_0^{-1}}}=\frac{N_{\mathrm{eff}}(E_F,H)}{N_0(E_F)} \tag{2}$$

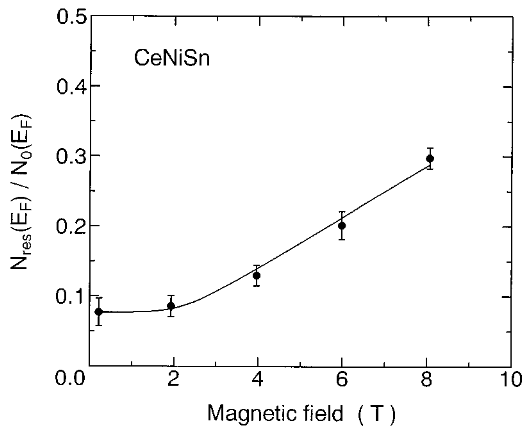

FIG. 3. Magnetic field dependence of the DOS fraction, $N_{\mathrm{res}}(E_F)/N_0(E_F)$. The solid line indicates the calculation based on the simple model that the V-shaped DOS is split by the magnetic field as illustrated in Fig. 1(b).

at very low temperatures where $N_{\mathrm{eff}}(E_F,H) = \sqrt{N_+(E_F-gJ\mu_BH)N_-(E_F+gJ\mu_BH)}$ and $(T_1T)_0^{-1}$ is estimated from the DOS, and $N_0(E_F)$ at E_F for the quasiparticle band with Lorentzian shape.[10,11]

The DOS fraction $N_{\mathrm{eff}}(E_F,H)/N_0(E_F)$ estimated from the above relation to be 0.077, 0.086, 0.130, 0.202, and 0.298 is plotted in Fig. 3 against magnetic fields, $H=0.22$, 1.93, 4, 6, and 8 T, respectively. The solid line is the calculation mentioned above, being in good accordance with the experiment. The DOS plateau with $N_{\mathrm{eff}}(E)/N_0(E)=0.077$ inside the pseudogap in Fig. 1(a) is distributed over the respective energy scale in the temperature (K) and the magnetic field (T) equal to about $\sim$ 2 K and $\sim$ 2 T. Accordingly, provided that the relative energy shift, $\Delta(k_B)=2gJ\mu_BH$ is smaller than $\sim$ 2 T for the up- (N_+) and the down-(N_-) spin bands split by the magnetic field, $N_{\mathrm{eff}}(E_F,H)$ is expected to stay nearly constant. After the Fermi level crossing over the DOS plateau with further increasing magnetic field, $N_{\mathrm{eff}}(E_F,H)$ starts to increase significantly.

Recent magnetoresistance measurement, which has revealed two remarkable peaks around 2 and 11.4 T at 0.45 K along the a axis, seems to be consistent with the structure inside the gap.[9] The V-shaped DOS model as in Fig. 1(a) is qualitatively compatible with the theoretical model proposed by Ikeda and Miyake.[12] They have derived the pseudogap behavior with a finite DOS at the Fermi level by noting that the hybridization vanishes on points along the a axis, in case that the $|5/2,\pm 3/2\rangle$ state among three doublets of $J=5/2$ is dominantly hybridized with conduction electrons. Then it has been argued that this anisotropic hybridization model is in accord with the anisotropic magnetic and transport properties observed in CeNiSn. It is noteworthy that a weak semimetallic behavior in CeNiSn arises through the weak k dependence of the f-electron self-energy.[12]

In conclusion, it has been found that $1/T_1$ below 1 K is markedly enhanced when a magnetic field exceeds 2 T, and the $T_1T=$const relation becomes valid from higher temperature and its magnitude increases continuously as the magnetic field increases up to 8 T. From these results, it has been shown that the quasiparticle band splits into up- and down-spin bands with V-shaped pseudogap unchanged by applying the magnetic field and eventually the DOS increases at the Fermi level. By contrast, applying a stronger magnetic field than 8 T has been suggested to significantly affect the V-shaped pseudogap itself. The application of $H=8$ T is effectively equivalent to either holes or electrons doping with 1%. Taken together with other experiments, the present NMR study has provided important clues that CeNiSn possesses a unique semimetallic ground state with a V-shaped pseudogap, being consistent with previous NMR results on the carrier doping effect.[5]

We are grateful to Professor K. Miyake and H. Ikeda for valuable discussions. This work was supported by the Scientific Research Grant-in-Aid from the Ministry of Education, Science and Culture of Japan.

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