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Transport and Magnetic Properties of a New Heavy-Fermion Metamagnet $CeNi_2Ge_2$

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Recent experimental results in the heavy-fermion compound $CeNi_2Ge_2$ are presented. We have studied the temperature dependence of electrical resistivity, thermopower, magnetic susceptibility and high-field magnetization. A quadratic temperature dependence in the resistivity is observed below 0.7 K. The susceptibility exhibits a broad maximum at 28 K. The magnetization shows a metamagnetic-like behavior at $H_M \sim 42$ T as in $CeRu_2Si_2$. These results indicate that $CeNi_2Ge_2$ is a new heavy-fermion compound showing a metamagnetic-like transition in the non-magnetic ground-state.

KEYWORDS: $CeNi_2Ge_2$, heavy-fermion, Fermi-liquid, non-magnetic ground state, metamagnetism

§1. Introduction

In the physics of the non-magnetic heavy-fermion systems, it is important to understand the ground state properties. These properties have been studied in $CeRu_2Si_2$[1,2] and $CeCu_6$[3]. At room temperature, each Ce ion in these compounds has a localized moment and behaves as a Kondo impurity. As indicated by inelastic neutron scattering measurements,[4] antiferromagnetic correlations between the localized moments develop gradually with decreasing temperature. The correlation length increases rapidly below about the characteristic temperature which is of the order of the energy gap. However, the correlation length does not diverge but saturates to a finite value, which is due to the single-site fluctuations giving rise to the Kondo effect.

In the ground state, the conduction electrons are coupled with the lattice of the dynamically correlated f-moments. A quasi-particle band with a large effective mass is thought to result from the unknown coupling between f-electron lattice and the conduction electrons. Thus, the f electrons in these compounds possess two particular characters. One is that for the quasi-particle band electrons near the Fermi-energy, the other is that for the localized electrons at the deep f-level. In other words, the f-electrons in the ground state possess both the itinerant character of a valence fluctuation regime and the localized character of a Kondo regime. This dual nature of the ground state indicates whether these compounds are in the proximity of the boundary between these regimes. It is still unclear that the itinerant and localized characters of the f electrons cross over smoothly or discontinuously at the boundary.[5]

The metamagnetic-like behavior in magnetization is a remarkable feature of $CeRu_2Si_2$ and $CeCu_6$, which shows up roughly below the Kondo temperature, T_K. The pseudo-metamagnetic transition fields H_M are 7.7 T and 2 T for $CeRu_2Si_2$[1] and $CeCu_6$[6,7], respectively. In the past decade, much interest has been focused on their metamagnetic properties. Especially in $CeRu_2Si_2$, a variety of physical properties related to the metamagnetic-like transition have been investigated. These results are shown in a recent review paper by Flouquet et al.[8] It appears that the aspect of each property is well clarified by precise measurements. However, the link between these properties is not sufficiently understood and still a matter of controversy. For example, the dHvA experiments indicating the itinerant-localized transition of f-electrons at H_M[9] apparently contradict the magnetization measurements indicating the absence of first order transition at H_M.[10,11] To get a complete understanding of the mechanism of the metamagnetic transition, comparative studies with between other heavy-fermion metamagnetic compound are important. Since metamagnetic Ce compounds with non-magnetic ground state are still rare, a search for other metamagnetic compounds is needed.

We have directed our attention to $CeNi_2Ge_2$ with the tetragonal $ThCr_2Si_2$ type structure. This compound is a non-magnetic heavy-fermion compound. The series of CeT_2X_2 (T = transition metals and X = Si and Ge) with the $ThCr_2Si_2$ type structure show a variety of ground states. The magnetic behavior of Ce is sensitive to the Ce-T and Ce-X spacings.[12] For the homologs with T = Ru, Ni, Cu, Ag and Au,[13] magnetic ordering is observed in the compounds which have large Ce - T distances ($d_{Ce\text{-}T}$ > 3.25~3.3 Å). The valence-fluctuation state is observed for $d_{Ce\text{-}T}$ well below 3.2 Å. For the non-magnetic heavy-fermion compounds, $CeRu_2Si_2$,

$CeCu_2Si_2$ and $CeNi_2Ge_2$, every $d_{Ce\text{-}T}$ is near 3.2 Å. This fact implies that these compounds are in the boundary region between the magnetic and the non-magnetic behaviors. Thus, an analogous nature of ground states between $CeNi_2Ge_2$ and $CeRu_2Si_2$ is expected.

The magnetic properties of $CeNi_2Ge_2$ have been investigated by Knopp *et al.*[14] by AC susceptibility, specific heat and inelastic neutron scattering measurements. They confirmed by AC susceptibility measurements that there is no indication of either a superconducting transition or a magnetic ordering down to 30 mK. The specific heat coefficient, $\gamma = C/T$, amounts to 350 mJ/mol K^2. This value is of the same order as in $CeRu_2Si_2$.[2] The energy width of the quasi-elastic peak decreases with decreasing temperature and becomes nearly constant below 30 K. The existence of a large residual width implies that strong single-site fluctuations, due to the Kondo effect, exists down to the lowest temperature. The magnetic phase diagram of the pseudo ternary system $Ce(Ni_{1-x}Cu_x)_2Ge_2$ have been investigated by Loidl et al.[15] With increasing Cu concentration, heavy-fermion band magnetism with incommensurate ordering vector appears between x= 0.75 and 0.2. This behavior is similar to the weak antiferromagnetic order as observed in $Ce(Ru_{1-x}Rh_x)_2Si_2$ system.[16]

The physical properties of $CeNi_2Ge_2$ under high magnetic fields have been investigated by several groups so far. Magnetoresistance measurements were early performed up to 14 T by Kletowski *et al.*[17] Recently, the magnetization was measured up to 35 T by Loidl *et al.*[15] and up to 40 T by Matsuhira *et al.*[18] In these results, there is no anomaly precursory of a metamagnetic transition in the above field ranges. It seemed worth to investigate the magnetic properties in a higher field region.

We have grown large single crystals of $CeNi_2Ge_2$. We have studied magnetization in fields up to 50 T to search for a metamagnetic transition, in addition to electrical resistivity, thermopower and magnetic susceptibility measurements by using these crystals.

§2. Experimental

The single crystals of $CeNi_2Ge_2$ and $LaNi_2Ge_2$ were grown by the Czochralski pulling method using a tetra-arc furnace. The typical sizes of the crystals obtained are 4~5 mm in diameter and 30~40 mm in length. The pulling rate is usually lower than 10 mm/hour. In order to improve quality, as-grown single crystals were purified by the solid-state electro-transport (SSE) with a DC current of ~1000 A/cm^2 under a pressure of 5×10^{-10} torr for a week. From X-ray diffraction measurements, lattice constants were found for the *a*- and *c*-axes 4.150 Å and 9.842 Å, respectively.

The electrical resistivity and thermopower were measured by the conventional 4-probe and the differential methods, respectively. The magnetic susceptibility was measured with a SQUID magnetometer at 500 Oe. The high-field magnetization measurements with pulsed magnetic field up to 50 T were performed at the High Magnetic Field Laboratory, of the Research Center for Extreme Materials, Osaka University.

§3. Results and Discussions

3-1 Transport properties

This sub-section is devoted to detail the temperature dependence of the electrical resistivity and the thermopower in the single crystalline samples of $CeNi_2Ge_2$. It will be shown that the overall behaviors of these transport coefficients are similar to those observed in the typical non-magnetic heavy-fermion compounds $CeRu_2Si_2$[1,8] and $CeCu_6$[3], indicating that $CeNi_2Ge_2$ can be also classified among the non-magnetic heavy-fermion.

Figure 1 shows the temperature dependence of the electrical resistivity for the single crystalline samples of $CeNi_2Ge_2$ and $LaNi_2Ge_2$ for the current along the *c*-axis ($\rho_{//}$) and the *a*-axis ($\rho_{\perp}$). The resistivity is anisotropic. For $LaNi_2Ge_2$, $\rho_{\perp}$ is larger than $\rho_{//}$ below the room temperature. The same anisotropy magnitude is also observed in $LaRu_2Si_2$[19] and YRu_2Si_2 [20] which crystallize in the same $ThCr_2Si_2$ type of structure, suggesting characteristic anisotropy of the resistivity of this structure. In this $LaNi_2Ge_2$ specimen, dHvA oscillations have been observed.[21] This fact indicates the good quality of the sample. For $CeNi_2Ge_2$, the resistivity of single crystalline samples have been reported by Schneider et al.[22] The resistivity data in ref. 22 are considerably different from our data, especially for $\rho_{//}$. Although $\rho_{\perp}$ is consistent with our data, $\rho_{//}$ is 2~3 times larger than our data in whole. In our measurements, we have observed that $\rho_{//}$ is very sensitive to the quality of the crystal as compared to $\rho_{\perp}$. For example, we have confirmed that $\rho_{//}$ increases in whole by several times of heat-cycle between 300 K and 4.2 K. We suppose that this is associated with micro-cracks occurring in the *c*-plane which is a cleavage plane. Hence, we believe that the temperature dependence of the resistivity shown in Fig. 1 is intrinsic.

Fig. 2 shows an enlarged view of the resistivity below 8K. The residual resistivities are subtracted from the data. Below 0.7 K, a T^2 dependence of the resistivity is observed. This is a typical feature of Fermi-liquid systems. For typical

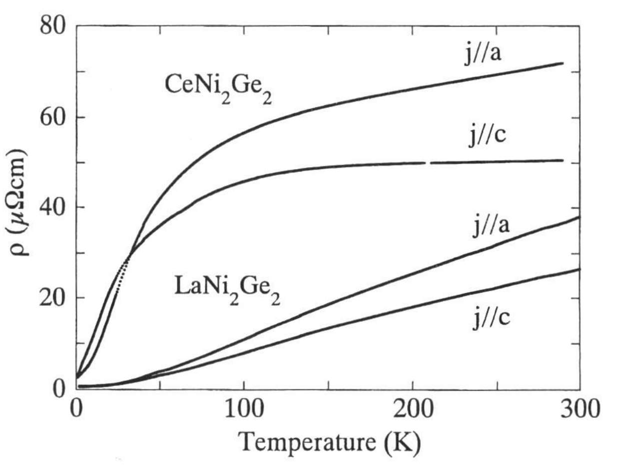

Fig. 1. Temperature dependence of electrical resistivity ρ of $CeNi_2Ge_2$ and $LaNi_2Ge_2$ single crystals.

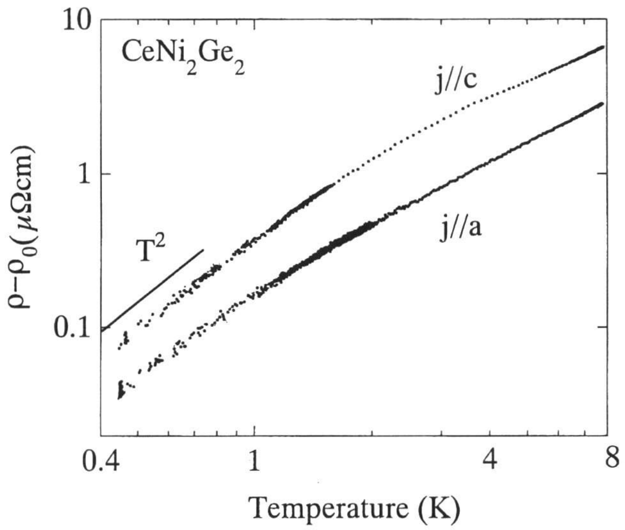

Fig. 2. The log-log plot of $\rho-\rho_0$ of $CeNi_2Ge_2$ single crystal as a function of temperature, where ρ_0 is the residual resistivity.

non-magnetic heavy fermion compounds, a T^2 dependence of the resistivity is empirically observed below a characteristic temperature, T_{SQR}, which roughly corresponds to $T_K/100$. For examples, T_{SQR} ~ 0.2 K[1,8] in $CeRu_2Si_2$ (T_K ~23 K) and T_{SQR} ~ 0.03 K[23] in $CeCu_6$ (T_K ~5 K). In $CeNi_2Ge_2$, T_K is reported to be 30 K[14]. This is roughly consistent with T_{SQR} ~ 0.7K. The coefficient of the T^2 term, A, is estimated to be 0.39 and 0.19 μΩcm/K^2 for the current along the c- and a-axes, respectively.

Next we show in Fig. 3 (a) the temperature dependence of the magnetic resistivity ρ_m, where $\rho_m = \rho_{CeNi_2Ge_2} - \rho_{LaNi_2Ge_2}$ and (b) the temperature dependence of the thermopower S for $CeNi_2Ge_2$. In Fig. 3(a), a magnetic resistivity shows a large positive peak at 100 K which are interpreted as the combined effect between the single ion-Kondo effect and the crystalline electric field (CEF), as is predicted by Cornut and Coqblin.[24] Similar behavior of the magnetic resistivity are observed also in $CeRu_2Si_2$[19] except that the temperature of the peak is 200 K in $CeRu_2Si_2$. At lower temperature, the magnetic resistivity of both $CeNi_2Ge_2$ and $CeRu_2Si_2$ show shoulders at around T_K.

The overall behavior of the thermopower in $CeNi_2Ge_2$ is consistent with those reported previously for single crystalline samples[22] and a polycrystalline sample[15]. The thermopower has a large positive peak at 100 K and a weak positive peak near 20 K. The origins of the structures in S of heavy-fermion compounds are still a matter of controversy. For $CeRu_2Si_2$ single crystals,[25] two positive peaks are observed in S: a broad positive peak near 230 K and small positive peak roughly near T_K ~ 23 K. Based on the results of thermopower measurements for La doped samples and under magnetic fields, the authors in ref. 25 interpreted that the origins of these peaks are (i) an interplay between incoherent Kondo scattering and CEF effect for the peak around 230 K and (ii) reminiscent of the Kondo resonance of each ion for the peak near 23 K. It appears that similar interpretations are applicable to the cases in $CeNi_2Ge_2$. To confirm this hypothesis, the determination of

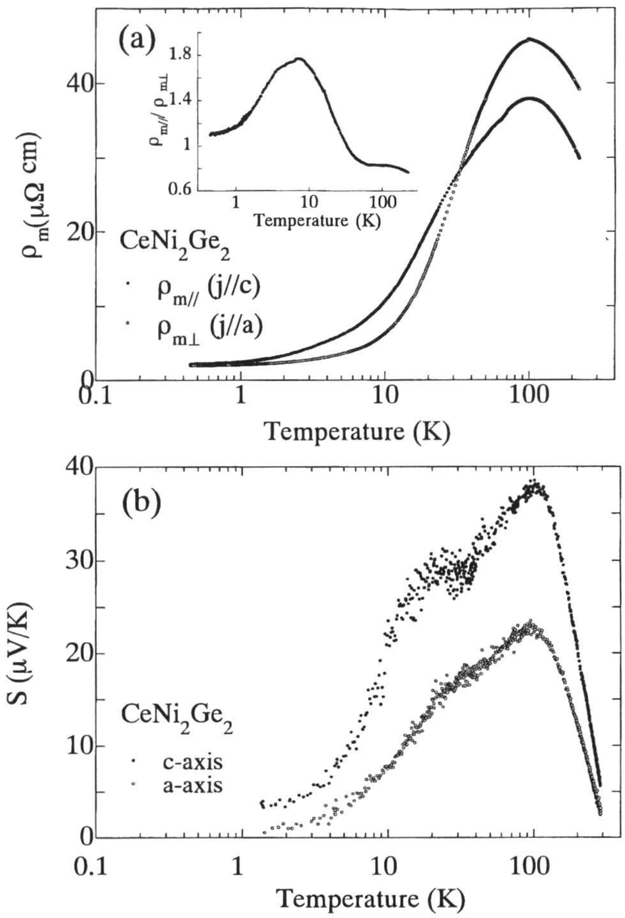

Fig. 3 (a) Temperature dependence of the magnetic resistivity ρ_m, where $\rho_m = \rho_{CeNi_2Ge_2} - \rho_{LaNi_2Ge_2}$. The inset show the anisotropy of the magnetic resistivity $\rho_m(j//c)/\rho_m(j//a)$. (b) Temperature dependence of the thermopower.

the CEF levels in $CeNi_2Ge_2$ should be performed. The double peak structure in S are observed also in single crystals of $CeCu_6$.[26] One is near T_K ~ 5 K and the other is around the splitting energy of CEF levels, ~ 60 K. These facts suggest that the double peak structure might be a characteristic behavior of S in non-magnetic heavy-fermion compounds.

The inset in Fig. 3 (a) shows the temperature dependence of the anisotropy of the magnetic resistivity, $\rho_{m//}/\rho_{m\perp}$. Evans et al. calculated the anisotropy of the magnetic resistivity in non-cubic materials taking into account both the crystal-field and Kondo effects.[27] The anisotropy of the magnetic resistivity generally possesses different values between that for the resistivity due to single-ion Kondo scattering ($T > T_K$) and that for the residual resistivity in the Fermi-liquid region ($T < T_K$).[26,27] In $CeNi_2Ge_2$, the anisotropy is roughly constant (≈ 0.8) between room temperature and 80 K, probably corresponding to the anisotropy due to the Kondo scattering. Near T_K (30 K) where the resistivity curves are crossing each other, the anisotropy changes strongly and is again constant below about 0.7 K. The constant value below 0.7 K corresponds to the anisotropy of the residual resistivity in the Fermi-liquid region. The overall behavior of the anisotropy is similar to that in $CeRu_2Si_2$[19], except for the magnitude of the anisotropy of the residual resistivity. We should note that the residual magnetic resistivity is quite sensitive to the accuracy of the measurements and the sample quality for both $CeNi_2Ge_2$ and $LaNi_2Ge_2$. For an accurate determination of the anisotropy of the residual resistivity, more precise measurements are needed. The anisotropy of A is estimated to: $A_{\perp}/A_{//}$,~ 0.5.

3-2 Magnetic properties

Figure 4 shows the temperature dependence of the magnetic susceptibility, χ, for the field along the *c*- and *a*-axes measured at 500 Oe. As indicated by the linear temperature dependence of χ^{-1} (the inset of Fig. 4), the susceptibilities for both directions obey the Curie-Weiss law, $\chi = C/(T+\theta)$, between 100 K and 300 K. The effective magnetic moments and the paramagnetic Curie temperatures are estimated to be 2.8 μ_B and 58 K for the *c*-axis, and 2.6 μ_B and 215 K for the *a*-axis. A broad maximum is found around $T_{\chi max}$ ~ 28 K for the field along the *c*-axis. This behavior is similar to that in $CeRu_2Si_2$, except that $T_{\chi max}$ is 10 K in $CeRu_2Si_2$. The origin of the upturn of the susceptibility at the lowest temperatures is not clear. The data in this region was fitted to a Curie law, assuming that this upturn is due to paramagnetic impurities. By subtracting this Curie component we could estimate the intrinsic susceptibility. In Fig. 4, the dotted curve shows the corrected susceptibility. In the corrected susceptibility, $T_{\chi max}$ is found around 30 K. Hence, we believe that $T_{\chi max}$ is not severely affected by the upturn at the lowest temperature. The origin of the upturn should be clarified by other measurements, such as Knight shift measurements.

Figure 5 shows the high-field magnetization of the free powder specimen in fields up to 50 T at 4.2 and 1.3 K. To secure enough penetration of the pulsed magnetic field, the measurements were performed on powder samples. The powder specimen was prepared with the single crystals purified by the SSE method. A metamagnetic-like behavior is observed at very high fields, with a clear peak in the differential susceptibility dM/dH as can be seen in the inset of Fig. 5. The critical field H_M, defined as the peak position in dM/dH, is 42.0 T at 1.3K and 42.8 T at 4.2 K. It is remarkable that H_M in $CeNi_2Ge_2$ is quite higher than that in $CeRu_2Si_2$ (H_M = 7.7 T). We note that the magnitude of the magnetization below H_M corresponds to an average value between the *a*- and *c*-axes magnetization. Namely, no crystal reorientation occurs in the fields. Although the magnetic anisotropy is weak as indicated by the absence of crystal reorientation, we believe that the present metamagnetic behavior occurs for the field along the *c*-axis as in $CeRu_2Si_2$. This is because the maximum of the paramagnetic susceptibility which is closely related to the pseudo-metamagnetic transition, is observed only for the field along the *c*-axis for $CeNi_2Ge_2$, as in $CeRu_2Si_2$[1])

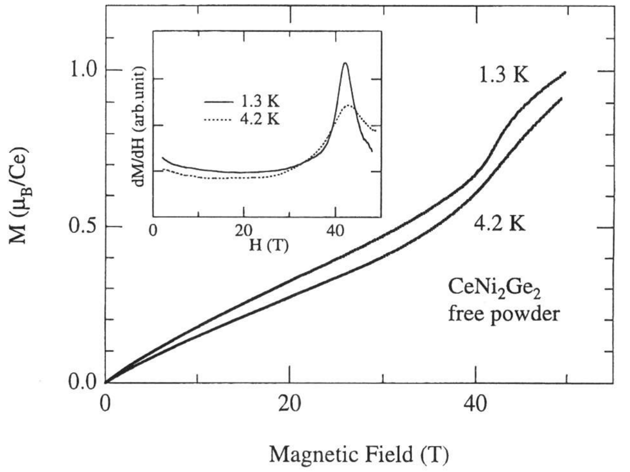

Fig. 5 Field dependence of the magnetization M of $CeNi_2Ge_2$ free powder. The inset shows the differential magnetization dM/dH.

In the following we wish to show the relation between H_M and some characteristic energies in $CeRu_2Si_2$, $CeCu_6$ and $CeNi_2Ge_2$. We will discuss the significant parameter in the heavy-fermion pseudo-metamagnetic transitions.

Figure 6 (a) shows a plot of H_M *vs.* $\Gamma_{s\text{-}s}$, where $\Gamma_{s\text{-}s}$ indicates the residual energy width of the magnetic peak in the quasi-elastic neutron scattering. Since $\Gamma_{s\text{-}s}$ relates to the energy-scale of the single-site fluctuations, this corresponds to the single ion-Kondo temperature T_K. In this plot, we have obtained $\Gamma_{s\text{-}s}$ for $CeNi_2Ge_2$ from ref. 14; $\Gamma_{s\text{-}s}$ for $CeRu_2Si_2$ and $CeCu_6$ from ref. 4; H_M of $CeRu_2Si_2$ and $CeCu_6$ from refs. 1 and 6, respectively. Apparently, the proportionality does not hold between H_M and $\Gamma_{s\text{-}s}$. This fact indicates that the pseudo-metamagnetic transition field might not scale by the single-ion Kondo temperature.

Next we show H_M *vs.* $T_{\chi max}$ plot in Fig. 6 (b), data points for $Ce_{0.95}La_{0.05}Ru_2Si_2$[28]) and $CeRu_2Si_2$ at pressures of 0, 1, 2, 4 and 5.8 kbar[29]) are also shown for comparison. It appears that a proportionality does not hold between H_M and $T_{\chi max}$. This fact suggests that the more than two parameters are significant to determine H_M and $T_{\chi max}$ for these compounds. It is remarkable that rather universal proportionality holds between H_M and $T_{\chi max}$ for the metamagnetic transition

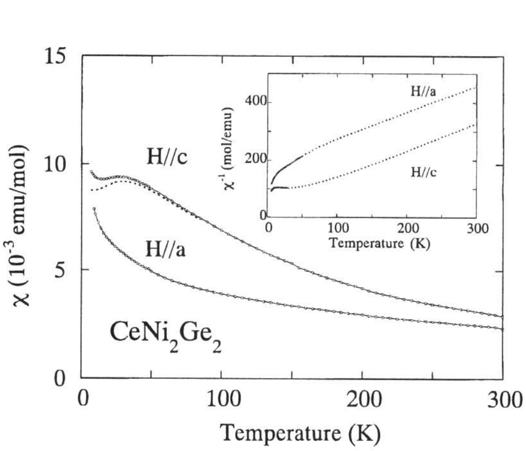

Fig. 4 Temperature dependence of the magnetic susceptibility χ of $CeNi_2Ge_2$ single crystal. The dotted line is the corrected susceptibility χ_c; $\chi_c = \chi - C_0/T$. The Curie constant C_0 in the correction term is estimated from the fitting below 10 K. The inset shows the inverse of the susceptibility.

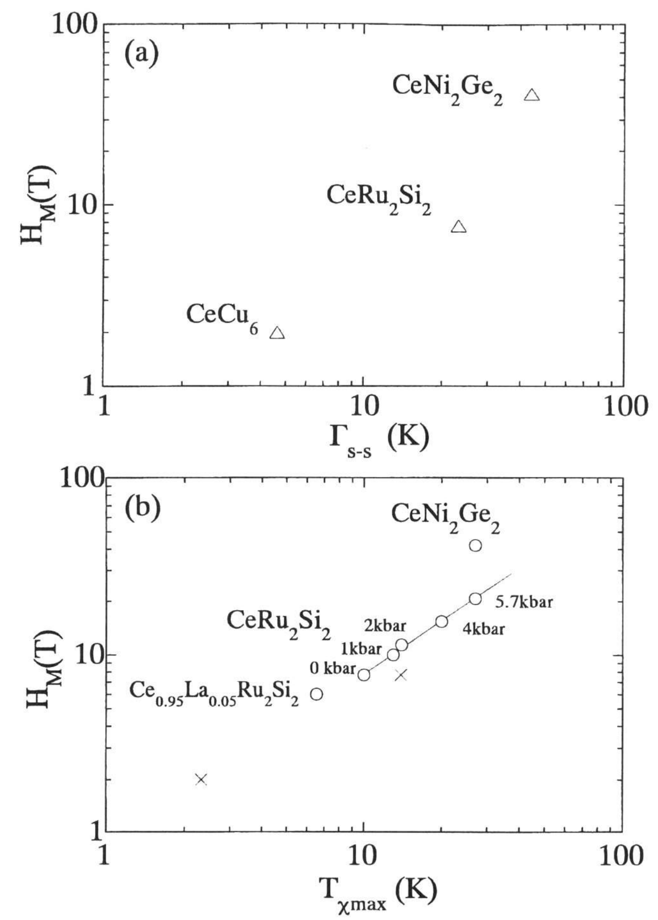

Fig. 6 (a) Pseudo-metamagnetic transition field H_M versus the residual width of the quasi-elastic scattering peak of the neutron diffraction. (b) Pseudo-metamagnetic transition field H_M versus the temperature of magnetic susceptibility maximum $T_{\chi max}$ in $CeNi_2Ge_2$, $Ce_{0.95}La_{0.05}Ru_2Si_2$[28] and $CeRu_2Si_2$ at pressures of 0, 1, 2, 4 and 5.6 kbar[29]. The solid line indicates the change of H_M and $T_{\chi max}$ in $CeRu_2Si_2$ by applying pressure. The crosses (X) show H_M *vs.* $\hbar\omega_0$ (in temperature unit of the horizontal-axis) plot for $CeCu_6$ (left) and $CeRu_2Si_2$ (right).

for the nearly ferromagnetic metals, such as $R(Co_{1-x}Al_x)_2$; R= Y and Lu.[30] This fact suggests the different nature of the metamagnetism between the heavy-fermion metamagnetism and the itinerant electron metamagnetism in the nearly ferromagnetic metals.

With regard to the parameter determining H_M, Rossat-Mignod et al.[4] suggest that the Zeeman energy corresponding to H_M is the order of the excitation energy, $\hbar\omega_0$, of the antiferromagnetic correlations. This value is determined by the energy transfer of the inelastic peak due to the antiferromagnetic correlations. In Fig. 6(b), H_M *vs.* $\hbar\omega_0$ (in temperature unit of the horizontal-axis) plot for $CeRu_2Si_2$ and $CeCu_6$ are shown by "X". If $\hbar\omega_0$ is the parameter determining H_M, it is expected that an inelastic peak in $CeNi_2Ge_2$ will be observed around an energy transfer of 6~8 meV (70 ~ 100 K). To elucidate the parameter relating the metamagnetic-like transition, inelastic neutron scattering measurements on single crystals of $CeNi_2Ge_2$ are needed. Also the investigation of the origin of $T_{\chi max}$, is important.

Summary

We have prepared single crystalline samples of $CeNi_2Ge_2$. These single crystalline samples were purified by the SSE method. They show typical features of transport and magnetic properties of non-magnetic heavy-fermion compound, as follows.

(1) T^2 dependence of the resistivity is observed below T_{SQR} ~ 0.7 K. This is roughly consistent with the empirical relation in non-magnetic heavy-fermion compounds.

(2) The thermopower shows two structures: a large positive maximum around 100 K and a weak positive peak near 20 K. This double peak structure is probably a characteristic behavior of S in non-magnetic heavy-fermion compounds.

(3) The magnetic susceptibility shows a maximum near 30 K for the field along to the c-axis.

(4) A metamagnetic-like behavior is observed in the magnetization curve at 4.2 and 1.5 K. The pseudo-metamagnetic transition field is 42.0 T at 1.5 K. This is a new example of heavy-fermion metamagnetism.

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