Pressure-induced magnetic instability in Pd-Ni alloys

Gendo Oomi1,2, Sadanori Iwai2, Masashi Ohashi3, Tomohito Nakano4
1) Department of Education and Creation Engineering, Kurume Institute of Technology, Kurume, Fukuoka 830-0052, Japan
2) Department of Physics, Kyushu University, Fukuoka 812-8581, Japan
3) Faculty of Environmental Design, Kanazawa University, Kanazawa 920-1192, Japan
4) Faculty of Engineering, Niigata University, Niigata 950-2181, Japan

E-mail: geomi@cc.kurume-it.ac.jp

Abstract. The electrical resistivity \( \rho(T) \) of dilute Pd-Ni alloys has been measured at high pressure up to 3 GPa. It is found that the \( \rho(T) \) of the ferromagnetic Pd-Ni alloy shows an anomalous temperature dependence near the critical pressure \( P_c \), where the ferromagnetism disappears. The results are analysed in the framework of quantum critical behaviour induced by pressure. The effect of magnetic field on the \( \rho(T) \) is also examined. Different behaviour of magnetoresistance against pressure was found depending on the alloy concentration and discussed in connection with an instability of ferromagnetism.

1. Introduction
There have been a lot of investigations of the quantum phase transition (QPT) for the intermetallic compounds including 4f or 5f rare earth elements, which is usually induced by changing control parameters such as concentration, magnetic field and pressure [1,2]. However a few reports on the QPT have been reported for the compounds or alloys with 3d transition metal elements [3,4]. It has been well known that Pd metal is an exchange-enhanced Pauli paramagnet and a ferromagnetism is easily induced by a small addition of magnetic elements such as Ni or Fe. A lot of experimental and theoretical investigations have been carried out for dilute Pd-Ni alloys [5,6]. Recently Nicklas et al. [7] and Aliev et al. [8] reported that non Fermi liquid (NFL) behaviors were observed near the critical concentration of \( x_c \) = 0.025 in dilute Pd\(_{1-x}\)Ni\(_x\) alloys and near the critical saturation field \( H_s \) in Fe/Cr magnetic multilayers [8]. In their results characteristic temperature dependent electrical resistivity \( \rho(T) \) was observed such as \( T^n \) with \( n = 1-1.5 \), which is largely different from the normal Fermi liquid (FL) behavior of \( n = 2 \). In order to get a deep insight into the new mechanism of QPT, it is worthwhile to explore the QPT in the wide range of materials. In the present work, the electrical resistivity \( \rho(T) \) of dilute Pd\(_{1-x}\)Ni\(_x\) alloys near the critical concentration of \( x_c \) has been measured at high pressure up to 3 GPa and high magnetic field. The \( \rho(T) \)s are analysed by assuming the relation \( \rho(T) = \rho_0 + A'T^n \), where \( \rho_0 \) is the residual resistivity, \( A' \) and \( n \) are constants.

2. Experimental
The polycrystalline samples of Pd\(_{1-x}\)Ni\(_x\) alloys near \( x_c \) were prepared by arc melting the constituent metals of 6N Pd and 4N Ni. The alloys were annealed at 1000 °C for 5 days in an evacuated silica tube. High pressure was generated by using a piston-cylinder method up to 3 GPa. The electrical resistance
was measured by means of standard four-probe method. The magnetic field was generated by using a superconducting magnet. The details of the high pressure apparatus were described previously[9].

3. Results and discussion

3.1. Effect of pressure on the temperature dependent electrical resistivity $\rho(T)$ of Pd-Ni alloys

We have measured the $\rho(T)$ of three Pd$_{1-x}$Ni$_x$ alloys with $x = 0.03, 0.027$ and $0.025$. In the following we indicate first the results for $x = 0.027$. Figure 1 illustrates the $\rho(T)$ at various pressures. All $\rho(T)$ curves show smooth temperature dependence without any crossing and discontinuities below room temperature. The pressure derivative of resistance is $-5.2 \times 10^{-2}$ GPa$^{-1}$ at room temperature, which is much larger than those of Fe and Cr [10].

![Figure 1](image1.png)

**Figure 1.** $\rho(T)$ of $x = 0.027$ at various pressures.

The $\rho(T)$ at low temperature was fit to the following equation, $\rho(T) = \rho_0 + A'T^n$. Figure 2 shows the values of $n$ and $A'$ as a function of pressure. It is found that the $n$ ($\approx 1.75$ at ambient pressure) decreases with increasing pressure below 1 GPa, shows a minimum near 1.5 GPa ($= P_c$) with $n$ around 1.6, and increases above 1.8 GPa. Since the $n$ is 2 for normal FL, the present result indicates that the electronic state of $x = 0.027$ deviates significantly from the normal FL, particularly around $P_c$. Furthermore the values of $A'$ are found to show a maximum around $P_c$. Recently such behaviours have been observed for several intermetallic heavy Fermion (HF) compounds including f elements such as cerium and is called as non Fermi liquid (NFL) property[11,12]. The behaviours mentioned above are considered as a direct evidence of quantum critical point (QCP) and often discussed in connection with the occurrence of superconductivity. It was reported for CePtSi$_2$ that the values of $n$ have a minimum near 1.4 GPa ($n \approx 1$) and also the residual resistivity shows a maximum associated with superconductivity around $T_c = 0.12$ K[13].

On the other hand there have been a few reports for the QCP behavior in the alloys or compounds including d elements. The NFL behaviour for Pd$_{1-x}$Ni$_x$ alloys has been reported [7,14] around $x_c (= 0.025)$, where $n$ shows a minimum and $A'$ has a maximum. In the present work, we have observed the same behaviour around $P_c$ in $x=0.027$ alloy. Thus the present results as shown in Fig.2 indicate that the alloy shows a quantum critical behaviour at high pressure near $P_c$. From the temperature dependence of magnetic susceptibility of this alloy, the Curie temperature $T_C$ is estimated to be about 5 K [7]. Since the $T_C$ of ferromagnetic $x = 0.03$ ($T_C \approx 15$ K) decreases with increasing pressure having the rate -3K/GPa, the $T_C$ of $x = 0.027$ may disappear around 1.7 GPa assuming the same pressure coefficient, which is in good agreement with $P_c$ of $x = 0.027$. 

![Figure 2](image2.png)

**Figure 2.** Temperature exponent $n$ and the coefficient $A'$ as a function of pressure.
Next we show the results for other concentrations, $x = 0.03$ and $0.025$. The $x = 0.025$ alloy is paramagnetic, which is just on the critical concentration $x_c$, and shows a NFL behaviour even at ambient pressure. In fact, the $\rho(T)$ shows a temperature dependence with $n \approx 1.5$ at ambient pressure. On the other hand, since the $n$ of ferromagnetic $x = 0.03$ is 1.9 at ambient pressure, the electronic state is described by normal FL. The pressure dependence of $n$ is shown in Fig. 3 for these two alloys. It is found that $n$ decreases with increasing pressure for ferromagnetic $x = 0.03$ and smoothly increases for paramagnetic $x = 0.025$. These results imply that as pressure increases, both the FL state of $x = 0.03$ and the NFL state of $x = 0.025$ become unstable but the change in the magnitude of $n$ of $x = 0.027$ by applying pressure is larger than that of $x = 0.03$. Considering that the $n$ of $x = 0.025$ approaches 2 as pressure increases, the FL state of $x = 0.025$ is expected to recover above 3 GPa.

The values of $A'$ are shown for $x = 0.03$ and $0.025$ in Figure 4 as a function of pressure. There is a large difference among two alloys. $A'$ increases smoothly with pressure for $x = 0.03$ but decreases significantly for $x = 0.025$. Since the ferromagnetic state of $x = 0.03$ is stable at ambient pressure, the change of $A'$ is relatively small. But the change of $A'$ of $x = 0.025$ is larger than that of $x = 0.03$. This result implies that a magnetic quantum fluctuation in $x = 0.025$ is larger than that in $x = 0.03$. As the large fluctuation in $x = 0.025$ near QPT is suppressed by applying pressure, the FL state recovers at high pressure above 3 GPa.

3.2. Effect of magnetic field on the electronic state of $x=0.027$.

Figure 5 indicates the $n$ as a function of magnetic fields $B(T)$ at 1.4 GPa, i.e., near QPT. $n$ increases rapidly below 0.3 T but above 1 T, the increasing rate tends to saturate and $n$ is 1.9 at 2 T, which is the same as that of $x = 0.03$ showing normal FL at ambient pressure. In other words the NFL state at $B=0$ becomes unstable by applying magnetic field and the FL state recovers around 2T. Since the electronic state of $x = 0.027$ is considered to be dominated by large quantum fluctuation due to magnetic instability around 1.4 GPa, the present result indicates that this fluctuation is suppressed by applying magnetic field. This observation is consistent with the previous results[15]. Furthermore the $A'$ at 1.4 GPa decreases with increasing $B$ from 1.8 nΩcm/K$^2$ at 0 T to 0.5 nΩcm/K$^2$ at 2 T, which is also nearly the same as that of $x = 0.03$ at ambient pressure (FL state).

Figure 6 shows the magnetoresistance (MR) ratio=$((\rho(H)-\rho(H = 0))/\rho(H = 0))$ at $T = 4.2$ K and $B = 6$ T for the present alloys as a function of pressure. Positive MR ratios of 20-30% were found for the all samples. But the pressure dependence is different each other. The MR ratio of $x = 0.025$ decreases smoothly with pressure, but that of $x = 0.027$ decreases slightly below 1.5 GPa but above it a small discontinuous drop of about 2% is observed.
The MR ratio of \( x = 0.03 \) is almost constant below 3 GPa reflecting relatively stable ferromagnetism compared with \( x = 0.027 \) alloy. The large pressure dependence of \( x = 0.025 \) is due to the large quantum fluctuations. Since the MR ratio depends strongly on the characteristic temperature such as spin fluctuation or Kondo temperature, \textit{i.e.}, the magnitude of \( A' \)[16], the small drop near \( P_c \) may be related to the QCP.

The authors would like to express their sincere thanks to Dr. M. J. Nicklas for his useful suggestions and showing them his PhD thesis.

References

[1] Y. Uwatoko \textit{et al.}, The Handbook on the Physics and Chemistry of Rare Earths, vol.42 (to be published).
[2] see for example, Proc. of Novel Pressure-induced Phenomena in Condensed Matter Systems, ed. by T.Kagayama, M. Ohashi and Y.Uwatoko, J. Phys. Soc. Jpn, Suppl. A, 76 (2007).
[3] C. Thessieu, C. Pfleiderer, and J. Flouquet, Physica B, 239 (1997) 67.
[4] G. Oomi, Y. Fuchizaki, M. Ohashi,T. Nakano and Y. Uwatoko, J. Magn. Soc. Jpn. 34 (2010) 263.
[5] R. A. Beyerlein and D. Lazarus, Phys. Rev. B, 7 (1973) 511.
[6] T. Kato and J. Mathon, J. Phys. F:Metal Phys., 6 (1976) 221.
[7] M. Nicklas, M. Brando, G. Knebel, F. Mayr, W. Trinkl, and A. Loidl, Phys. Rev. Lett., 82 (1999) 4268.
[8] F. G. Aliev, V. V. Moshchalkov, and Y. Bruynseraede, Phys. Rev. Lett., 81 (1998) 5884.
[9] F. Honda, S. Kaji, M. Ohashi, G. Oomi, T. Eto and T. Kagayama, J. Phys. :Conden. Matter, 14 (2002)11501.
[10] P. W. Bridgman, The Physics of High Pressure, (Dover, New York,1970) 264.
[11]G. R. Stewart, Rev. Mod. Phys. 73 (2001) 797.
[12] H. Miyagawa, G. Oomi, M. Ohashi, I. Satoh, T. Komatsubara, M. Hedo and Y. Uwatoko, Phys. Rev. B, 78 (2008) 064403.
[13] T. Nakano, M. Ohashi, G. Oomi, K. Matsubayashi and Y. Uwatoko, Phys. Rev. B, 79 (2009) 172507.
[14] A. Tari and B. R. Coles, J. Phys. F: Metal Physics, 1 (1971) L69.
[15] M. J. Nicklas, PhD thesis, Univ. Augsburg, 2000.
[16] T. Kagayama, G. Oomi, R. Yagi, Y. Iye, Y. Onuki and T. Komatsubara, J. Phys. Soc. Jpn. 61 (1992) 2632.