Available online at www.sciencedirect.com
ScienceDirect
Journal of Magnetism and Magnetic Materials 310 (2007) 590–592
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

^{73}Ge-NQR study of heavy-fermion compound $CeNi_2Ge_2$

S. Kawasaki[a,*,1], T. Sada[a], T. Miyoshi[b], H. Kotegawa[b], H. Mukuda[a], Y. Kitaoka[a], T.C. Kobayashi[b], T. Fukuhara[c], K. Maezawa[c], K.M. Itoh[d], E.E. Haller[e]

[a]Department of Materials Engineering Science, Graduate School of Engineering Science, Osaka University, Toyonaka, Osaka 560-8531, Japan
[b]Department of Physics, Faculty of Science, Okayama University, Okayama 700-8530, Japan
[c]Faculty of Engineering, Toyama Prefectural University, Toyama 939-03, Japan
[d]Department of Applied Physics and Physico-Informatics, Keio University, Yokohama 223-8522, Japan
[e]Department of Materials Science and Engineering, University of California at Berkeley and Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

Available online 2 November 2006

Abstract

We report ^{73}Ge-nuclear-quadrupole resonance (NQR) study of heavy-fermion compound $CeNi_2Ge_2$. The temperature dependence of the ^{73}Ge nuclear-spin-lattice-relaxation rate $1/T_1$ indicates the development of magnetic correlations and the formation of a Fermi-liquid state at temperatures lower than $T_{FL} = 0.4\,K$, where $1/T_1T$ is constant. The $1/T_1T$ decrease below $T_c^{NQR} = 0.1\,K$, whereas resistance decreases below $T_c^{onset} = 0.2\,K$ and does not become zero. These results indicate $CeNi_2Ge_2$ closely locates to a superconducting quantum critical point.

PACS: 71.27.+a; 72.15.−v; 74.70.Tx; 76.60.Gv

Keywords: Heavy fermion; $CeNi_2Ge_2$; NQR

Unconventional superconductivity(SC) observed around the antiferromagnetic (AFM) quantum critical point (QCP) has been one of the most important issues in cerium (Ce)-based heavy-fermion (HF) compounds, since it was universally found at the border of antiferromagnetism [1] .

The HF compound $CeNi_2Ge_2$ crystallizes in the $ThCr_2Si_2$ structure. The measurements of resistivity and specific heat at low temperatures clearly revealed non-Fermi-liquid-like behaviors associated with antiferromagnetism [2–4]. So, $CeNi_2Ge_2$ is suggested to locate near the AFM QCP. Remarkably, there exist several reports indicating that resistance becomes zero below $T_c{\sim}0.2\,K$ in $CeNi_2Ge_2$, suggesting the onset of SC [3,5]. Therefore, it is suggested that magnetic fluctuations with regard to the AFM QCP are responsible for the onset of SC in this compound.

Here, we report ^{73}Ge nuclear quadrupole resonance (NQR) measurements on $CeNi_2Ge_2$. High-quality single crystals of ^{73}Ge-enriched $CeNi_{2.02}Ge_2$ were grown by the Czochralski method and moderately crushed into grains in order to enable easy penetration of the rf pulses into the samples. However, to avoid crystal distortions, the size of the grains is kept larger than 100 μm. Note that the onset of SC in this compound is extremely sensitive to the sample preparation method and/or the nominal stoichiometry in the Ni element. The sharpest SC transition was reported for the Ni-rich sample $CeNi_{2.02}Ge_2$ [5]. The T dependences of ^{73}Ge-NQR spectra and $1/T_1$ were measured at $H = 0$ down to $T = 0.03\,K$ using a ^{3}He/^{4}He dilution refrigerator. In order to detect the possible onset of some magnetic ordering at low temperatures, the NQR spectrum for the $1\nu_Q$ transition was precisely measured by the Fourier transform method for spin-echo signals.

Fig. 1 shows the temperature dependence of $1\nu_Q$ transition of ^{73}Ge-NQR spectra for $CeNi_2Ge_2$ with $\nu_Q = 1.632\,MHz$ and the asymmetry parameter $\eta = 0$ due to the uniaxial symmetry. Note that the full width at half

*Corresponding author. Tel./fax: +81 6 6850 6438.
E-mail address: kawasaki@science.okayama-u.ac.jp (S. Kawasaki).
[1]Present address: Department of Physics, Faculty of Science, Okayama University, Okayama 700-8530, Japan.

0304-8853/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.jmmm.2006.10.232

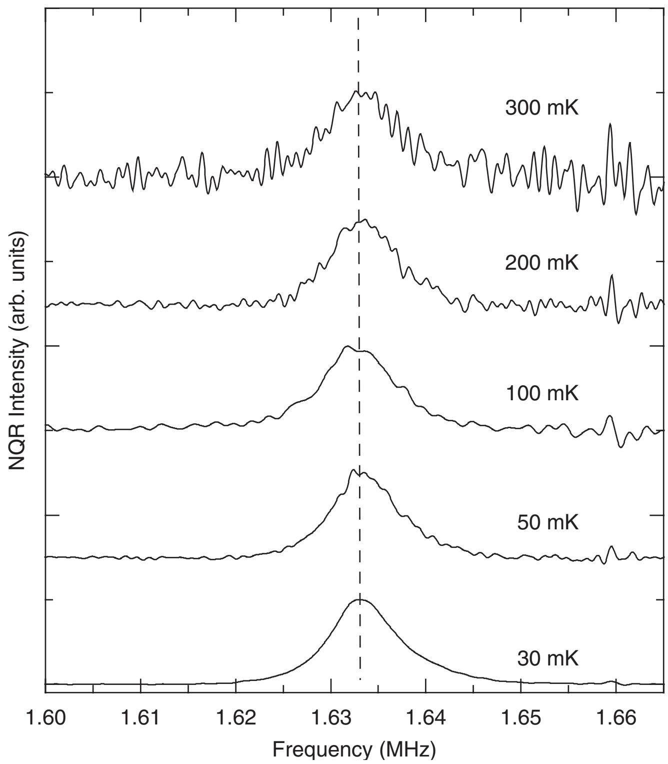

Fig. 1. Temperature dependence of the ^{73}Ge $1\nu_Q$-NQR spectrum in $CeNi_2Ge_2$. Dotted line denotes the peak position.

maximum (FWHM) for the $1\nu_Q$-NQR spectrum is quite sharp at FWHM $= 8$ kHz, confirming the high quality of the sample used in this study. Here, the NQR spectra can indicate the emergence of a static magnetic order for $CeNi_2Ge_2$ from the splitting and/or the broadening of the $1\nu_Q$-NQR spectrum due to the appearance of an internal field, if any. Since the FWHM of the $1\nu_Q$-NQR spectrum does not exhibit any change as shown in Fig. 1, there is no evidence for a magnetic order and/or a structural change down to 0.03 K in $CeNi_2Ge_2$.

Next, we present the T dependence of $1/T_1$ in $CeNi_2Ge_2$. Fig. 2 shows the T dependence of $1/T_1T$ under zero field ($H = 0$) (circle), $H = 0.03$ T (triangle), and $H = 0.1$ T (square) in the T range of $0.04-150$ K. The $1/T_1T$ at $H = 0$ increases as the temperature decreases to 0.4 K, thus revealing the growth of magnetic correlations. This result demonstrates that $CeNi_2Ge_2$ is very closely located to the AFM QCP. On the other hand, at temperatures lower than $T_{FL} = 0.4$ K, $1/T_1T$ is constant; this indicates the formation of the Fermi-liquid state.

A remarkable finding in the Fermi-liquid regime is that the $1/T_1T$ at $H = 0$ slightly decreases below $T_c^{NQR} = 0.1$ K. Note that this temperature is lower than $T_c^{onset} = 0.2$ K below which the resistance begins to decrease as shown in the inset of Fig. 2. In order to confirm whether this decrease in $1/T_1T$ is associated with the onset of SC, $1/T_1T$ was measured under external fields of $H = 0.03$ and 0.1 T, as shown by the solid triangles and open squares in the figure, respectively. The application of such tiny fields causes $1/T_1T$ to remain constant down to

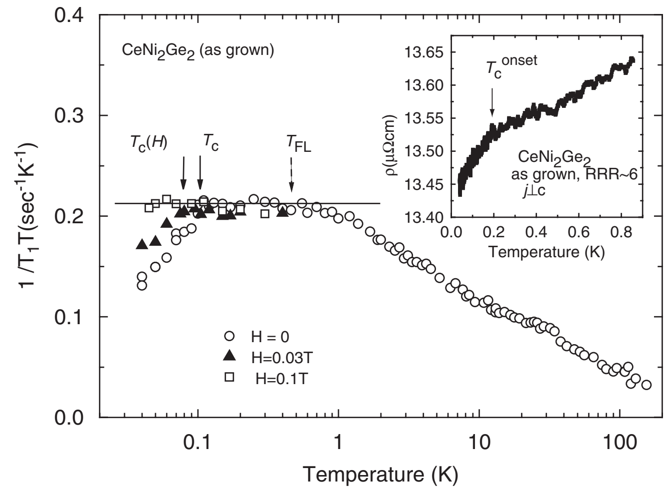

Fig. 2. Temperature dependence of $1/T_1T$ for $CeNi_2Ge_2$ (open circle). Solid triangles and open squares indicate the temperature dependence of $1/T_1T$ under a magnetic field of $H = 0.03$ and 0.1 T, respectively. Solid line indicates a $1/T_1T =$ const. law. Solid and dashed arrows indicate T_c and $T_c(H)$ and T_{FL} (see the text), respectively. Inset shows the temperature dependence of resistivity. Solid arrow indicates the T_c^{onset}.

$T = 0.07$ and 0.04 K at $H = 0.03$ and 0.1 T, respectively. This result possibly provides evidence for the onset of SC at $T_c^{NQR} = 0.1$ and 0.07 K at $H = 0$ and 0.03 T, respectively. Since $1/T_1T$ indicates a power-law like dependence without a coherence peak just below T_c, the origin of SC in $CeNi_2Ge_2$ seems to be unconventional.

Notably, $1/T_1T =$ const. behavior is observed down to 0.04 K for the annealed sample. Although the sample quality is improved by annealing, this SC anomaly in $1/T_1T$ as well as resistance disappears [6]. This is possibly because a slight unconventional SC in $CeNi_2Ge_2$ is observed in the heavy Fermi-liquid state in the close vicinity of the AFM QCP. As a result, the annealed sample seems to be apart from the superconducting QCP.

In conclusion, systematic studies of ^{73}Ge NQR in $CeNi_2Ge_2$ have revealed the development of magnetic correlations down to 0.4 K followed by the formation of a Fermi-liquid state below $T_{FL} = 0.4$ K. A remarkable finding is that the significant reduction in $1/T_1T$ is associated with the onset of SC below $T_c = 0.1$ K, this is because the application of a tiny magnetic field causes $1/T_1T$ to remain constant without a reduction of T_c below 0.1 K. These results are due to the emergence of the unconventional superconducting fluctuations in association with the superconducting QCP. This is because zero resistance is not observed. As a result, these results are consistent with the fact that bulk SC does not emerge but the superconducting coherence length remains finite over a short-range distance due to the closeness to the superconducting QCP.

This work was partially supported by a Grant-in-Aid for Creative Scientific Research (15GS0213) from the Ministry of Education, Culture, Sports, Science and Technology

(MEXT) and the 21st Century COE Program (G18) by Japan Society of the Promotion of Science (JSPS).

References

[1] Y. Kitaoka, et al., J. Phys. Soc. Japan 74 (2005) 186 and references therein.
[2] F. Steglich, et al., J. Phys. Condens. Matter 8 (1996) 9909.
[3] F.M. Grosche, et al., cond-mat/9812133 (1998).
[4] Y. Aoki, et al., J. Phys. Soc. Japan 66 (1997) 2993.
[5] P. Gegenwart, et al., Physica B 281 & 282 (2000) 5.
[6] S. Kawasaki, et al., J. Phys. Soc. Japan 75 (2006) 043702.