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Pressure-induced antiferromagnetic superconductivity in $CeNiGe_3$: A ^{73}Ge-NQR study under pressure

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Abstract

We report on antiferromagnetic (AF) properties of pressure-induced superconductivity in $CeNiGe_3$ via the ^{73}Ge nuclear-quadrupole-resonance (NQR) measurements under pressure (P). The NQR-spectrum measurements have revealed that the incommensurate antiferromagnetic ordering is robust against increasing P with the increase of ordered moment and ordering temperature. Nevertheless the measurements of nuclear spin-lattice relaxation rate ($1/T_1$) have pointed to the onset of superconductivity as a consequence of Ce-4f electrons delocalized by applying P. The emergence of superconductivity under the development of AF order suggests that a novel type of superconducting mechanism works in this compound.

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The pressure-induced superconductivity (SC) was recently discovered in an antiferromagnetic (AF) compound $CeNiGe_3$ with a Neel temperature $T_N = 5.5$ K under pressure (P) [1]. Two SC domes (SC1 and SC2) were found in the P ranges of 1.9–3.5 GPa and 5.9–7.3 GPa, respectively [2]. T_N increases as P increases up to $P = 3$ GPa and disappears around $P \sim 7$ GPa. Interestingly, the SC1 phase coexisting with the AF phase exists far from a quantum critical point (QCP) at $P \sim 7$ GPa, while the SC2 phase emerges near the QCP as well as in Ce-based heavy-fermion superconductors such as $CeCu_2Si_2$ [3], $CeCu_2Ge_2$ [4], $CePd_2Si_2$ [5], $CeRh_2Si_2$ [6], $CeIn_3$ [7] and $CeRhIn_5$ [8]. Another remarkable point is that the superconducting transition temperature T_{sc} in the SC1 dome exhibits a peak around $P \sim 3$ GPa where the Neel temperature does also with $T_N \sim 8.5$ K. These results suggest a novel type of superconducting mechanism which differs from the previous examples. Here we report on the characteristics of the AF phase where the SC1 takes place via the ^{73}Ge nuclear-quadrupole-resonance (NQR) studies above $T = 1.4$ K at $P = 0$, 2.0 and 2.8 GPa.

A ^{73}Ge-enriched polycrystalline sample was prepared and crushed into powder to make oscillating magnetic field penetrate into the sample easily. ^{73}Ge-NQR spectra shown in Fig. 1(a) are well separated with peaks arising from three inequivalent Ge sites in a unit cell. Each Ge site is assigned comparing with the values of quadrupole frequency on the basis of the band calculation performed by Harima et al. as shown in Table 1. We note that a small amount of other Ce–Ni–Ge compounds are actually contained in the sample, but each NQR spectral width for $CeNiGe_3$ is narrow enough with a full width at half maximum (FWHM) being ~40 kHz, assuring good quality of the

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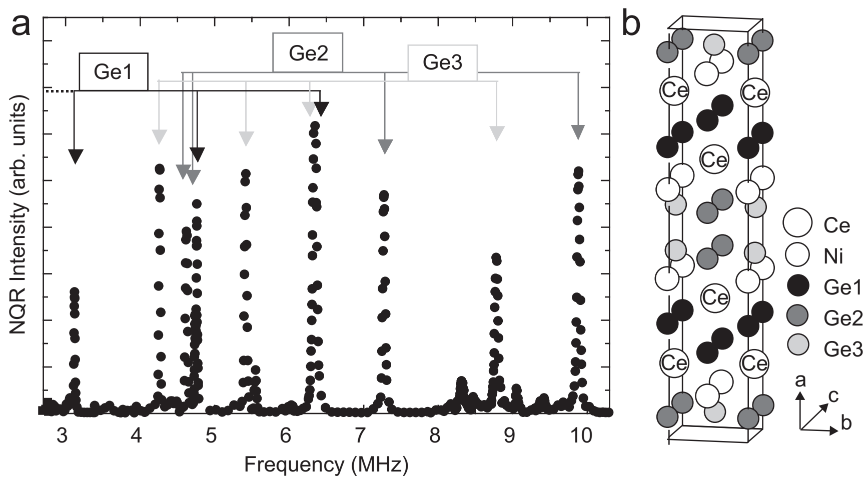

Fig. 1. (a) NQR spectra for the paramagnetic state at $T = 10\,\mathrm{K}$ and $P = 0$, which are separated into peaks associated with the three inequivalent Ge sites in a unit cell. (b) Crystal structure of $CeNiGe_3$.

Table 1
NQR parameters (ν_Q, η) at Ge(1), Ge(2) and Ge(3) sites, which are estimated by the experiment (Exp.) and band calculation (Cal.), respectively, at $P = 0\,\mathrm{GPa}$

Ge(1)	ν_Q (MHz)	η	Ge(2)	ν_Q (MHz)	η	Ge(3)	ν_Q (MHz)	η
Exp.	1.59	0.11	Exp.	2.51	0.40	Exp.	2.57	0.56
Cal.	1.481	0.475	Cal.	2.200	0.388	Cal.	2.041	0.561

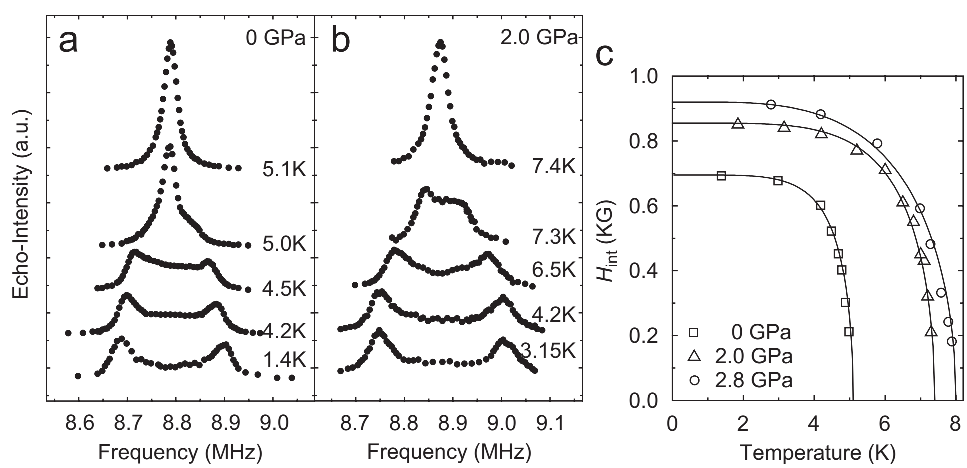

Fig. 2. Temperature dependence of NQR spectra at (a) $P = 0$ and (b) 2.0 GPa. Spectral shapes reveal the onset of the incommensurate AF ordering below $T_N = 5.1$ and 7.4 K, respectively. (c) The temperature dependence of internal magnetic field at $P = 0$ (open squares), 2.0 (open triangles) and 2.8 GPa (open circles), at Ge(3) site. The solid curves are the calculation based on a mean field approximation.

sample. As temperature (T) decreases below T_N, the spectra shown in Figs. 2(a) and 2(b) reveal a pattern ascribed to a distribution of internal field H_{int} in magnitude and/or direction, which are consistent with the occurrence of an incommensurate AF order (IAF) below $T_N = 5.1$, 7.4 and 8.0 K at $P = 0$, 2.0 and 2.8 GPa, respectively. Fig. 2(c) indicates the T dependence of H_{int} at the Ge(3) site which is scaled to that of an ordered moment M_{IAF}. The increase of H_{int} is due to the development of M_{IAF} below T_N under P (see Fig. 2(c)). Here, $H_{int}(T = 0) = 0.70$, 0.86 and 0.92 kG are estimated at $P = 0$, 2.0 and 2.8 GPa, respectively. As T_N increases, $M_{IAF} \propto H_{int}\,(T = 0)$ also increases. These results indicate that the IAF is robust against applying P, exhibiting the increase of T_N and M_{IAF} in$CeNiGe_3$. Even in a situation such that T_N increases, it is remarkable that SC1 sets in, which has been never reported so far.

Fig. 3 shows the T dependence of $1/T_1$ at $P = 0$, 2.0 and 2.8 GPa at the Ge(1) site. $1/T_1$ above T_N stays almost constant at $P = 0\,\mathrm{GPa}$, suggesting that Ce-4f electrons-derived magnetic moments behave as almost localized at

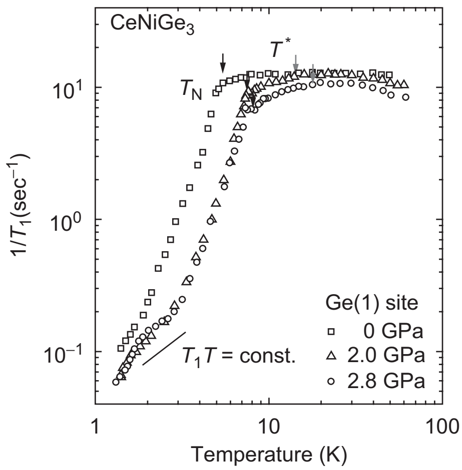

Fig. 3. Temperature dependence of $1/T_1$ at $P=0$ (open squares), 2.0 (open triangles) and 2.8 GPa (open circles), at the Ge(1) site. The black and gray arrows indicate T_N and T^*, respectively.

ambient P. On the other hand, the respective $1/T_1$'s at $P=2.0$ and 2.8 GPa start to decrease below $T^*\sim 14$ and 18 K that are significantly larger than $T_N = 7.4$ and 8.0 K, probing an increase in the hybridization between 4f-electrons and conduction electrons which makes 4f electrons mobile via forming heavy electrons. As T^*, below which 4f electrons tend to be itinerant, increases with P, T_N and $M_{AF} \propto H_{int}$ increase, stabilizing the AF order in $CeNiGe_3$. The presence of density of states at the Fermi level is confirmed by the observation of a $T_1T = \text{const.}$ behavior in the AF state around $T = 2.5$ K at $P = 2.0$ and 2.8 GPa. The $T_1T = \text{const.}$, that becomes valid in a narrow T range, starts to decrease below $T\sim 2$ K lower than T_N. At the same temperature $T\sim 2$ K, the nuclear spin–spin relaxation rate ($1/T_2$) undergoes a peak, suggesting a development of longitudinal fluctuations of ordered moment in the IAF state. These results suggest that an unusual evolution towards a different kind of ordered phase may take place under P. Further experiments to address the novel nature in the IAF are required in the low T region and under high P.

In summary, the ^{73}Ge-NQR measurements in $CeNiGe_3$ above $T = 1.4$ K at $P = 0$, 2.0 and 2.8 GPa have revealed that the incommensurate antiferromagnetic order is robust against increasing P with the increase of ordered moment and ordering temperature. Nevertheless the measurements of nuclear spin-lattice relaxation rate ($1/T_1$) have pointed to the onset of superconductivity as a consequence of Ce-4f electrons delocalized by applying P. The emergence of superconductivity under the development of AF ordered state suggests that a novel type of superconducting mechanism works in this compound.

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