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Intimate interplay between superconductivity and antiferromagnetism in CeNiGe₃: A ⁷³Ge-NQR study under pressure

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Abstract

We report the ⁷³Ge-NQR studies on the antiferromagnetic (AFM) heavy-fermion compound CeNiGe₃ which shows two domes like pressure-induced superconducting phases in the pressure (*P*) ranges of 1.7–3.7 GPa and 5.9–7.3 GPa denoted as SC1 and SC2, respectively [M. Nakashima, et al., J. Phys. Condens. Matter. 16 (2004) L255, H. Kotegawa, et al., J. Phys. Soc. Jpn. 75 (2006) 044713]. The NQR spectra have revealed a change from an incommensurate AFM structure at P = 0 and 2.0 GPa into a commensurate one at P = 2.8 GPa. The onset of the SC1 may be relevant to an intimate evolution from the incommensurate into commensurate AFM-spin structure as *P* increases.

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The heavy-fermion antiferromagnet CeNiGe₃ ($T_N = 5.5 \text{ K}$) becomes superconducting under high pressure (P) [1]. Most spectacular is that the pressure-induced superconductivity emerges in two domes in the P ranges of 1.7–3.7 GPa and 5.9–7.3 GPa denoted as SC1 and SC2, respectively [2]. An application of pressure (P) makes T_N increase. As shown in Fig. 1(a) [2,3] T_N exhibits a maximum around $P\sim3$ GPa, and disappears around $P\sim7$ GPa. It should be noted that SC1 appears under a deep inside of antiferromagnetic (AFM) phase, while SC2 emerges around the quantum critical point (QCP) where the AFM order collapses as well as the case for the Cebased P-induced superconductors such as CeCu₂Si₂ [4], CeIn₃ [5], CeRhIn₅ [6], etc. The emergence of SC1 may be due to Ce-4f electrons delocalized, even though the AFM order is robust against by the application of P which makes $T_{\rm N}$ and internal magnetic field increase [3]. These experimental results suggest a novel type of superconducting mechanism in SC1 which differs from SC2 near QCP. Here we report on the characteristics of the P dependence of AFM structure revealed by the ⁷³Ge nuclear-quadrupole-resonance (NQR) studies under P, and discuss about the relationship between the antiferromagnetism and the SC1 in CeNiGe₃.

The ⁷³Ge-enriched polycrystalline sample was prepared and crushed into powder to allow maximal penetration of oscillating magnetic field. Hydrostatic pressure was applied by using piston cylinder cell filled with polyethylsiloxane as a pressure-transmitting medium in this study.

Figs. 2(a)–(c) show the respective NQR spectra for $4v_Q \ (\pm 7/2 \leftrightarrow \pm 9/2)$ transition at the Ge3 site in paramagnetic (PM) and AFM state at P = 0, 2.0, and 2.8 GPa. In the AFM state, an internal field (H_{int}) associated with an

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Fig. 1. (a) Schematic P-T phase diagram of CeNiGe₃ derived by the resistivity measurements [2]. The superconducting transition temperature T_{sc} for the SC1 and SC2 phases are determined from a temperature at which the resistance becomes zero as shown by the solid lines. The arrows point the *P* values of 0, 2.0, and 2.8 GPa. (b) Crystal structure of CeNiGe₃.



Fig. 2. NQR spectra in paramagnetic (PM) and antiferromagnetic (AFM) state well below T_N at (a) P = 0, (b) 2.0 GPa, and (c) 2.8 GPa, at Ge3 site. The solid line for the spectrum at P = 2.8 GPa is a simulation which assumes a unique value of H_{int} with a Lorentzian spectral shape as expected for the commensurate spin structure of AFM order below T_N . Note that the spectra at P = 0 and 2.0 GPa are not the case. (d) Schematic energy level for nuclear spin I = 9/2 in PM and AFM state.



Fig. 3. T_N (solid circles) and H_{int} at the Ge3 site (open squares) determined by NQR spectra at P = 0, 2.0, and 2.8 GPa. They are plotted in the P-T phase diagram of CeNiGe₃ derived by the resistivity measurements [2].

onset of AFM order causes the Zeeman splitting in the whole NOR spectra, making the NOR spectra split, as shown in Fig. 2(d). The shape and splitting in NOR spectra allow us to determine a possible spin structure and a size of ordered moment. We have determined the $T_{\rm N}$ and $H_{\rm int}$ from NQR spectra at each P, as shown in Fig. 3. Here we consider that the direction of H_{int} is parallel to the principal axis of the electric field gradient (V_{77}) at the Ge3 site, which is revealed by NQR spectra, and the band calculation performed by H. Harima indicates V_{zz} is parallel to b axis at the Ge3 site. The P dependence of $T_{\rm N}$ is consistent with the previous report performed by resistivity [2]. In addition to $T_{\rm N}$, the remarkable increase of the H_{int} is seen with application of P. It is interesting that the SC1 occurs under such a robust AFM state, which suggests the intimate interplay between superconductivity in SC1 and antiferromagnetism. Another important point is that the NQR spectrum well below $T_{\rm N}$ at $P = 2.8 \,{\rm GPa}$ consists of two Lorentzian spectra, pointing to the presence of a unique value of H_{int} at the Ge3 site. This is consistent with a commensurate structure of AFM order. By contrast

to the spectra at 2.8 GPa, the NQR spectra at P = 0 and 2.0 GPa exhibit a distribution in H_{int} , suggesting a possible change in spin structure. If a spin structure were of an incommensurate type exhibiting either a helical structure or spin-density-wave (SDW), a possible distribution in H_{int} would be expected at the Ge site as observed for the spectra at P = 0 and 2.0 GPa. The onset of the SC1 may be relevant with an intimate evolution from the incommensurate AFM structure as *P* increases. This is a contrast to the case for CeRhIn₅ where the superconductivity emerges under the incommensurate AFM state [8]. Further detailed *P* and *T* dependence of NQR spectra in AFM state are needed to investigate an interplay between the onset of SC1 and the change of spin structure as the function of *P*.

In summary, the ⁷³Ge-NQR measurements in CeNiGe₃ have revealed that the spin structure of AFM order evolves from the incommensurate to commensurate one with increasing P up to P = 2.8 GPa where SC1 emerges. We propose that the evolution into the commensurate AFM structure is relevant with the onset of the SC1 in CeNiGe₃.

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