Co-NQR Study on Successive Magnetic Phase under Pressure in Non-centrosymmetric CeCoGe$_3$

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Abstract. We have performed Co-NQR study to clarify the complex magnetic phases in approaching to the quantum critical point (QCP). The successive transitions at $T_{N2}=12$ K and $T_{N3}=8$ K after the ferrimagnetic-like order at $T_{N1}=21$ K in ambient pressure are confirmed by the spectral changes of Co-NQR. However no critical slowing down of the nuclear spin-lattice relaxation rate $1/T_1$ or the nuclear spin-spin relaxation rate $1/T_2$ at $T_{N2}$ and $T_{N3}$ was observed, suggesting 1st order transitions. In applying pressure, a large spectral change of Co-NQR occurs in relatively low pressure of 0.3 Gpa. The Co-NQR spectrum becomes simple above about 0.7 GPa, consisting of the two Co sites with spectral weight ratio of 2. No successive transitions were observed in 1.5 GPa, indicating that the successive transitions are confined to relatively low pressure region. The extremely slower decrease of the sublattice magnetization than that expected in the mean field approximation is seen in 1.5 GPa.

1. Introduction

Non-centrosymmetric CeCoGe$_3$ exhibits a complex magnetic order in a way to approach to the quantum critical point (QCP) around about 5.5 GPa.[1, 2] Below about 0.6 K around QCP, the heavy Fermion superconductivity without inversion symmetry appears.[3] Much attention have been payed to this system because of the Cooper pairing symmetry of the superconductivity in addition to the complex magnetic order despite of the only one crystallographic Ce site. We have performed Co-NQR study to clarify the the complex magnetic phase, and moreover to clarify the Cooper pairing symmetry in the non-centrosymmetric heavy Fermion superconductors as a final goal. In an ambient pressure, CeCoGe$_3$ shows a ferrimagnetic like order at $T_{N1}=21$ K, and the successive transitions at $T_{N2}=12$ K and $T_{N3}=8$ K with very strong anisotropic magnetization along easy c-axis and their step-like decrease at these temperatures.[4, 5] We have reported previously that these successive transitions are also verified microscopically.[6] The Co-NQR spectrum consists of the four components, four Co sites acquiring different internal field $H_{int}$ all parallel to the c-axis. The transitions at $T_{N2}$ and $T_{N3}$ are seen as the the change of the number and the relative weight of the spectral components. In this paper, we will report the dynamical aspect of the Ce moments in CeCoGe$_3$, the nuclear spin-lattice relaxation rate $1/T_1$ and the nuclear spin-spin decay rate $1/T_2$ in ambient pressure. After these, the NQR results under pressure will be reported focusing on the development of the successive transitions.
2. Experiments
The sample preparations and the pressure determination were described in the previous report.[6] NQR measurements were performed using the phase coherent spin echo method. The corrections for echo intensity, so called $\nu^2$ and the spin echo decay are made. $1/T_1$ is evaluated by the inversion-recovery method of the nuclear magnetization for the peak at the highest NQR frequency $3\nu_Q$ corresponding to the $(\pm 5/2 - \pm 7/2)$ transition. $1/T_2$ is evaluated by the decay of echo intensity with increasing the time interval between exciting and refocusing pulses.

3. Experimental results and discussions

![Figure 1. T-dependence of $1/T_1$ in CeCoGe$_3$ (○) at $T>T_{N1}$ and LaCoGe$_3$ (□). The closed circle represents the contribution (1/$T_1$)$_f$. The dotted line denotes the expectation based upon the localized moment model (see text).](image1)

![Figure 2. T-dependence of $1/T_2^*$ in CeCoGe$_3$. $T_2^*$ is the time until the nuclear magnetization decays to 1/e. The measurements performed at respective NQR peaks below $T_{N1}$.](image2)

Figure 1 shows $T$-dependence of $1/T_1$ and $1/T_2$ in ambient pressure. $1/T_1$ in the reference compound LaCoGe$_3$ follows the Korringa law ($T_1 T=0.932$ (secK)), indicating that $d$-electrons of Co atoms contribute to conduction band. Nearly the same value of $1/T_1$ in CeCoGe$_3$ as that in LaCoGe$_3$ above about 200 K implies that Co atoms in CeCoGe$_3$ retain no localized moment. The $T$-dependence of the $f$-electron contribution to the relaxation $(1/T_1)_f$, $(1/T_1)_f=(1/T_1)_{CeCoGe3}-(1/T_1)_{LaCoGe3}$, is interpreted in terms of the localized moment model of Ce 4f moment at least at higher temperatures, as $(1/T_1)_f \propto \gamma N_1 A_{hf}^2 \tau_f$, where $\tau_f$ is the characteristic fluctuation time of the 4f localized moment, $A_{hf}$ the hyperfine coupling strength between f electron and Co nuclear spin, $\gamma$ the gyromagnetic ratio of Co nucleus. Considering two mechanisms for $\tau_f$ as $(1/\tau)_f=(1/\tau)_{ff}+(1/\tau)_{fc}$, where $(1/\tau)_{ff}$ is the exchange fluctuation frequency through effective exchange coupling between 4$f$ electron spins, so that $(1/\tau)_{ff} \sim \omega_{ex}$ ($T$-independent), while $(1/\tau)_{fc}$ is the Korringa-like relaxation rate of 4f electron spin, then $(1/\tau)_{fc} \propto J^2 c_\rho (c_F)^3 k_B T \propto T$. Combining the two contributions, the $T$-dependence of $1/T_1$ is written as $1/T_1 \propto 1/(a + b T)$, with the two constant parameters, $a$ and $b$. The dotted line is a fit to this relation. The downward deviation below about 50 K might be ascribed to the crystalline field effect (first excitation energy of about 100 K) or Kondo effect, consistent with the resistivity measurement.[4]
Figure 3. (a) $P$-dependence of $H_{int}^{(i)}$ for respective Co sites. The inset shows the NQR spectra associated with $3\nu_Q + \gamma_N H_{int}^{(i)}$ lines under respective pressure. (b) $P$-dependence of the relative spectral weight for respective Co sites. The dotted lines correspond to 2/3 and 1/3.

The spin-spin decay rate $1/T_2$ exhibits a critical divergence at $T_{N1}$, indicative of 2nd order phase transition, however those at $T_{N2}$ or $T_{N3}$ do not show any changes. The successive transitions are evidenced to be classified not to 2nd order but 1st order one. These are consistent with NQR results, abrupt changes of $H_{int}$ as well as the spectral weight, and also consistent with the recent neutron scattering measurements.[7]

Summarizing the present NQR results including the previous one, we get a image for the complex magnetic phases as, (1) the magnetism in this compound is owing only to Ce 4f localized moment, (2) Ising-like Ce moment along $c$-axis constructs more than two magnetic domains, producing $H_{int}^{(i)}$ ($i=1, 2, 3$ or 4), (3) the volume fraction of these domains as well as the magnetic structures change at $T_{N2}$ and $T_{N3}$ as a first order transitions.

Next we show the results under pressure. Figure 3 shows the pressure dependence of the NQR spectrum up to 1.52 GPa, the internal field and the integrated intensity measured at 4.2 K associated with the $3\nu_Q + \gamma_N H_{int}^{(i)}$ lines. The magnetic state corresponding to these results is regarded as the ground state because of $4.2 \text{ K} < T_{N3}$. However the NQR spectrum exhibits drastic change until above 0.7 GPa. The four Co sites reduces to two above about 0.3 GPa. As shown previously, in ambient pressure, four Co sites at $T < T_{N3}$ (phase III) and three Co sites at $T_{N2} < T < T_{N1}$ (phase I) are observed.[6] Thus the transition temperature $T_{N2}$ as well as $T_{N3}$ should decrease rapidly below at least 4.2 K with increasing pressure up to 0.3 GPa. Moreover the two Co sites above 0.7 GPa is not compatible with the three Co ones in the phase I. Therefore some vertical line in the $T$-$P$ phase diagram of the magnetic state should be required to discriminate the phase I and the new phase having two Co sites above 0.7 GPa. Corresponding to the NQR results, Mizoo et al shows that the spontaneous magnetization (existing prominently in the phase I) disappears, simultaneously its anomalous points with respect to temperature converge at 1.05 GPa.[2] Thus the magnetic structure giving two Co sites without net magnetization would be stabilized above about 1.0 GPa.

Figure 4 shows the $T$-dependence of the NQR spectrum under 1.52 GPa. The spectra for two Co sites (we denote A and B for high and low frequency peaks, respectively) decrease monotonously with increasing temperature, keeping the spectral weight ratio $(I_B/I_A)$ roughly constant of about 2. These behaviors indicate no successive transitions below $T_N$($\sim 19 \text{ K}$) any more. This is somewhat controversal to the $T$-$P$ phase diagram proposed by the specific
heat measurements. The characteristic step-like decrease of the transition temperature with increasing pressure is ascribed to the existence of stable magnetic structure corresponding to the respective steps of $T_N$, and these stable states are connected to the successive transitions in ambient pressure.[1] If it is true, the successive transitions would appear at around 12 K and 8 K at 1.52 GPa. However no evidences for the successive transitions were found.

We comment the magnetic structure possessing two Co sites with the weight ratio of 2. Here the moment directions within a $c$-plane, up and down, are denoted as $u$ and $d$, respectively. The $uuudd$ structure proposed as a basic magnetic structure from the neutron scattering in ambient pressure[8] provides the same number of 2 Co sites but with the different weight ratio of 1 if the internal field is assumed to transfer from the nearest neighbor and next nearest neighbor Ce moments. We notice that possible $uuuddd$ structure possessing smaller number of $ud$ pairs than those of $uu$ or $dd$ ones is compatible with the experimental spectral weight. Since NQR is a local probe, the long range magnetic structure cannot be predicted uniquely. The neutron scattering under pressure is highly desired.

Finally we mention the $T$-dependence of the sublattice magnetization under 1.52 GPa. Assuming that the sublattice magnetization is proportional to the internal field, $T$-dependence of the normalized internal field is shown in the inset of Fig. 4. The decrease of sublattice magnetization with increasing temperature is much slower than the mean field approximation in the case of $J=5/2$ presented by the solid line. Although the reason is not clear at present, the slow decrease is one of the characteristics of AF order under pressure.

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