NMR/NQR Study of pressure-induced superconductor CePt$_2$In$_7$

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Abstract. Zero-field $^{115}$In-nuclear magnetic resonance (NMR) has been used to study single crystals of CePt$_2$In$_7$, which is a heavy-fermion antiferromagnet with a Neél temperature ($T_N$) of 5.2 K at ambient pressure. The NMR spectra under zero field near the $Q$ line of the orthorhombic In(3) sites are taken at 1.6 K under hydrostatic conditions at ambient pressure and 2.4 GPa. These data reveal the coexistence of commensurate and incommensurate antiferromagnetic (AFM) orders at ambient pressure and that the commensurate ordering is stabilized by increasing pressures. The nuclear spin-lattice relaxation rates ($1/T_1$) for In(3) sites indicate the localized nature of $f$ electrons far above $T_N$. The values of $1/T_1$ in the paramagnetic state decrease by applying pressure. In contrast, the residual values of $1/T_1$ much below $T_N$ increase by pressure.

1. Introduction

Recently, an intermetallic compound CePt$_2$In$_7$ [1] has been added to the so-called Ce115 family. The Ce115 family members with a chemical formula of CeTIn$_5$ ($T$=Co, Rh, Ir) have attracted much attention since each is a heavy-fermion superconductor either at atmospheric pressure (CeCoIn$_5$ [2] and CeIrIn$_5$ [3]) or when antiferromagnetic (AFM) order is suppressed by applied pressure (CeRhIn$_5$ [4]). While the Ce115 structure (space group $P4/mmm$) can be regarded as an alternate stacking of CeIn$_3$ and TIn$_2$ layers along the $c$ axis, the structure of CePt$_2$In$_7$ ($I4/mmm$) is comprised of one CeIn$_3$ plane followed by two PtIn$_2$ layers stacked along the $c$ axis to form the tetragonal body-centered unit cell. Initial specific heat and electrical resistivity measurements on polycrystalline CePt$_2$In$_7$ showed that it is a heavy-fermion antiferromagnet with $T_N$ =5.2 K and that applying pressure induces a broad dome of superconductivity about the critical pressure $P_c$$\approx$ 3 GPa where $T_N(P_c)$ extrapolated to $T = 0$ [5]. Very recently, single crystals of CePt$_2$In$_7$ have become available, and like their polycrystal analogs, they order antiferromagnetically at $T_N$ = 5.2 K out of a heavy-fermion paramagnetic state [6]. Resistivity and $ac$-specific heat studies on these crystals as a function of pressure find a much narrower dome of pressure-induced superconductivity, even though the maximum $T_c$ and critical pressure $P_c$ are the same as in polycrystalline samples [7]. Compared to the Ce115, the dimensionality of CePt$_2$In$_7$ is reduced relative to the Ce115, as indicated by its crystal structure and band structure calculations [5] that are confirmed by quantum oscillation studies [8].
With its ability to study small single crystals, extreme sensitivity to lattice distortions and compatibility with high pressure environments, NQR/NMR is a powerful probe of the microscopic nature and pressure-induced evolution of magnetism in CePt$_2$In$_7$. Recently, we have reported the pressure and temperature dependences of $^{115}$In-NMR spectra under zero field for this material [9]. This experiment revealed the coexistence of commensurate (C-) and incommensurate (IC-) AFM orderings at ambient pressure and the stabilization of C-AFM state as the superconducting phase is approached with increasing pressure. In this brief paper, we report the characteristic changes of spectral shape and relaxation rates between ambient pressure and the hydrostatic pressure of 2.4 GPa.

2. Experimental

Single crystals for CePt$_2$In$_7$ were grown by a self-flux method. NQR and NMR measurements under zero field were performed by using a conventional phase-coherent pulsed NMR spectrometer in the temperature range 1.5-200 K. A dozen single crystals were mounted into an rf-coil in order to avoid decomposition and/or surface strain caused by crushing the crystals. It is noted that powdering or crushing crystals of CePt$_2$In$_7$ influences the physical properties, especially the volume fraction of IC-/C-AFM states [9]. Hydrostatic pressures up to 2.4 GPa were applied with a standard hybrid piston-cylinder-type clamp cell using Daphne7373 oil as a pressure medium.

3. Results and discussions

![Figure 1. Zero field NMR spectra under the ambient pressure and the hydrostatic pressure of 2.4 GPa at 1.6 K for In(3) sites in CePt$_2$In$_7$. The solid and dotted curves represent the simulated curves on the assumption of commensurate and incommensurate magnetic structures, respectively. The open triangles (▽) indicate the spectral positions for no internal field.]

All the nuclear quadrupolar resonance (NQR) lines for $^{115}$In were measured at ambient pressure in the paramagnetic state. These NQR lines can be ascribed to three crystallographically inequivalent In sites, that is, In(1) ($4/mmm$ in Hermann-Mauguin notation), In(2) ($4m2$), and In(3) ($2mm$). Because the overlap with other lines is minimal, the detailed spectral structure around $3Q$ of the In(3) sites is tracked with temperature and pressure [9]. Figure 1 shows the zero-field NMR spectra under ambient pressure and 2.4 GPa at 1.6 K for In(3) sites in CePt$_2$In$_7$. The spectrum at ambient pressure can be interpreted as coexistence of IC-AFM and C-AFM orders. The IC-AFM ordered portion provides a wing-shape spectrum (dotted curve in Fig. 1), and the C-AFM portion yields the remaining Gaussian-like peaks (solid curve in Fig. 1), including the peak at the same position as the NQR line determined above.
\( T_N \) (indicated by \( \Uparrow \) in Fig. 1). With applying pressures, the wing-shape spectrum gradually weakens until 2.4 GPa where the spectrum shows the Gaussian-like peaks. The simulations of these spectra are specified in Ref. [9]. The pressure dependence of the NMR spectra in zero field below \( T_N \) reveals the stabilization of C-AFM order as the superconducting phase is approached. It appears, therefore, that superconductivity in CePt\(_2\)In\(_7\) emerges as the C-AFM phase boundary is suppressed toward \( T = 0 \).

As noted above, the partly lingering NQR line implies the existence of sites partially canceling internal fields by the C-AFM arrangement. Indeed, as shown in Fig. 2, a sharp drop of nuclear spin-lattice relaxation rate \( 1/T_1 \) at \( T_N \) on this lingering line is observed, which reflects the emergence of an AFM energy gap. At the ambient pressure, the temperature dependence of \( 1/T_1 \) in the paramagnetic state above 25 K shows a constant behavior, which indicates the localized character of 4\( f \) electrons. Below \( T^* \approx 20 \text{ K} \), \( 1/T_1 \) starts to decrease gradually from the constant value. It suggests that an onset of \( c-f \) hybridization may occur around \( T^* \). As seen in Fig. 2, the critical slowing down just above \( T_N \) is observed over a quite narrow temperature window. Thus, \( T_N \) can be precisely determined by the \( 1/T_1 \) measurement.

The temperature dependence of \( 1/T_1 \) at 2.4 GPa is also shown in Fig. 2. At 2.4 GPa, \( T_N \) is determined to be 5.4 K from the peak of \( 1/T_1 \). This confirms microscopically that the AFM state is robust against pressures up to 2.4 GPa, which is consistent with the macroscopic measurements of resistivity and \( ac \)-specific heat [7]. In the paramagnetic state at 2.4 GPa, the values of \( 1/T_1 \) decrease from those at ambient pressure, and it does not show a constant value even around 100 K. By applying pressure, \( T^* \) seems to increase above 100 K, that is, 4\( f \) electrons procure itinerancy via \( c-f \) hybridization far above \( T_N \). Simultaneously, \( 1/T_1 \) shows a power-law temperature dependence of \( T^{1/4} \), which can be seen near an AFM instability. A similar temperature dependence of \( 1/T_1 \) has been observed in the well-known heavy fermion superconductor CeCoIn\(_5\) [10, 11]. On the other hand, the residual \( 1/T_1 \) after opening an AFM gap is found to increase with pressure. It may suggest an increase of residual density of states by mismatched nesting condition of Fermi surfaces, and/or persistent magnetic fluctuations even
below $T_N$. The origin of the residual $1/T_1$ under pressure may relate to the pressure-induced superconductivity in this material.

4. Conclusion
We have presented zero-field NMR spectra of the antiferromagnet CePt$_2$In$_7$, which indicates the coexistence of IC/C-AFM orderings at ambient pressure. The pressure dependence of the NMR spectra reveals the stabilization of C-AFM order as the superconducting phase is approached. The $1/T_1$ in the paramagnetic state under 2.4 GPa show a characteristic power-law temperature dependence near an antiferromagnetic instability. In order to understand the interplay among magnetism, quantum criticality, and superconductivity in CePt$_2$In$_7$, further NMR experiments under pressures are currently in progress.

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