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Published on: 23 May 2001 - Physical Review B (American Physical Society)

Pressure-induced anomalous magnetism and unconventional superconductivity in $CeRhIn_5$:^{115}In-NQR study under pressure

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(Dated: March 12, 2001)

We report ^{115}In nuclear-quadrupole-resonance (NQR) measurements of the pressure(P)-induced superconductor $CeRhIn_5$ in the antiferromagnetic (AF) and superconducting (SC) states. In the AF region, the internal field H_{int} at the In site is substantially reduced from $H_{int} = 1.75$ kOe at $P = 0$ to 0.39 kOe at $P = 1.23$ GPa, while the Néel temperature slightly changes with increasing P. This suggests that either the size in the ordered moment $M_Q(P)$ or the angle $\theta(P)$ between the direction of $M_Q(P)$ and the tetragonal c axis is extrapolated to zero at $P^* = 1.6 \pm 0.1$ GPa at which a bulk SC transition is no longer emergent. In the SC state at $P = 2.1$ GPa, the nuclear spin-lattice relaxation rate $^{115}(1/T_1)$ has revealed a T^3 dependence without the coherence peak just below T_c, giving evidence for the unconventional superconductivity. The dimensionality of the magnetic flutuations in the normal state are also discussed.

PACS numbers: PACS: 74.25.Ha, 74.62.Fj, 74.70.Tx, 75.30.Kz, 76.60.Gv

There is increasing evidence that a superconducting (SC) order in cerium (Ce)-based heavy-fermion (HF) compounds takes place nearby the border where an antiferromagnetic (AF) order is suppressed by applying pressure (P) to the HF-AF compounds $CeCu_2Ge_2$,[1] $CePd_2Si_2$[2] and $CeIn_3$.[3] When a magnetic medium is near an AF phase, AF waves of electron-spin density tend to propagate over a long distance with a low characteristic energy. Thereby it was argued that the binding of the Cooper pairs could be described in terms of the emission and absorption of fluctuating AF waves.[3] The interplay between the AF and SC states in the Ce-based HF systems may share some common aspects with other strongly-correlated-electron systems, and the understanding of mechanisms for superconductivity different from the conventional electron-phonon mediated one is still an important and unresolved issue.

Quite recently, it was discovered that a new HF-AF $CeRhIn_5$ (the Néel temperature, T_N=3.8 K) becomes a bulk HF superconductor at pressures exceeding $P_c \sim$1.63 GPa.[5] It was suggested that a first order-like transition from an AF state to a SC state occurs with a SC transition temperature $T_c \sim 2.2$ K that is nearly 10 times larger than the maximum values for $CePd_2Si_2$ and $CeIn_3$. Apparently, the evolution from the AF to SC states differs from all previous examples. T_N was reported to increase weakly with the pressure for $P \leq 1.45$ GPa, above which there is no resistive signature for T_N. In order to shed light on a P-induced exotic evolution from the AF to SC states in $CeRhIn_5$, one needs to uncover magnetic and SC characteristics through extensive experiments under P.

Here we report extensive ^{115}In-NQR measurements of $CeRhIn_5$ in the AF and SC states. The temperature (T) and P dependences of the ^{115}In-NQR spectrum and the nuclear spin-lattice relaxation rate $^{115}(1/T_1)$ were measured in a P range of $0-2.1$ GPa and a T range of $0.15-50$ K. The salient results are (1) T_N slightly increases up to P = 1.00GPa, but decreases at P=1.23 GPa, (2) By contrast, the internal field H_{int} at the In site due to the magnetic ordering is substantially reduced with P. This P-induced reduction in H_{int} might be attributed to either an ordered moment $M_Q(P) \to 0$ or the angle $\theta \to 0$ at $P^* = 1.6 \pm 0.1$GPa, where θ is the angle between the direction of $M_Q(P)$ and the tetragonal c axis, (3) The $^{115}(1/T_1)$ in the SC state at $P = 2.1$ GPa obeys a T^3 dependence without the coherence peak just below T_c consistent with a line-node gap model as reported in all previous HF-SC compounds, [4] (4) In the normal state at P=2.1GPa, the T dependence of $^{115}(1/T_1)$ is consistent with the three dimensional (3D) SCR theory for a nearly AF Fermi-liquid state.

Single crystal of $CeRhIn_5$ was grown by the self-flux method.[5] Powder X-ray diffraction indicated that the compound consists of a single phase that is formed in the primitive tetragonal $HoCoGa_5$ structure. The single crystal was moderately crushed into grains in order to make rf pulses penetrate into samples easily. Hydrostatic pressure was applied by utilizing a NiCrAl/BeCu piston-cylinder cell, filled with a Si-based organic liquid as a pressure-transmitting medium. High-frequency ac-susceptibility (ac-χ) was measured at P=1.5, 1.65, 1.8 and 2.15 GPa by using the *in-situ* NQR coil. The ac-χ data show a sharp SC transition at T_c=2.1 and 2.2 K at P=1.8 and 2.15 GPa, respectively (see Fig.4(b)). The ^{115}In-NQR spectrum was obtained by plotting spin-echo signal as a function of frequency in $T = 1.4-10$ K and $P = 0-2.1$ GPa. T_1 was measured by the conventional saturation-recovery method in $T = 0.15-50$ K at P=0, 1.23 and 2.1 GPa.

$CeRhIn_5$ consists of alternating layers of $CeIn_3$ and $RhIn_2$ and hence has two inequivalent In sites per a unit cell. The In(1) site, analogous to the single In site in a cubic $CeIn_3$, is located on the top and bottom faces of the tetragonal unit cell. By considering the symmetry of the In(1) site, this site was characterized by $\nu_Q = 6.78 \pm 0.01$ MHz and an asymmetry parameter η=0 in the previous NQR study.[6] Here ν_Q and η are defined by the NQR Hamiltonian: $H_Q = (h\nu_Q/6)[3I_z^2 - I^2 + \eta(I_x^2 - I_y^2)]$. The In(1)-NQR spectrum in the paramagnetic state at 4.2 K and $P = 0$ is shown in the bottom of Fig.1 where four transitions are found at the different frequencies $\nu = n\nu_Q$ with n=1, 2, 3 and 4, respectively.

In order to deduce the T dependence of the internal field $H_{int}(T)$ at the In(1) site in the AF state, we focus on the splitting of the $2\nu_Q$ (3/2$\Longleftrightarrow$1/2) transition and the shift of resonance frequency of the $3\nu_Q$ (5/2$\Longleftrightarrow$3/2) transition in the In(1) spectrum below T_N. This is because H_{int} lies perpendicular to the tetragonal c axis as reported in the previous study.[6] Including the Zeeman term $H_Z = -\gamma\hbar I_x \cdot H_{int}$ where γ is the gyromagnetic ratio, we diagonalize the full Hamiltonian $H_{nuc} = H_Q + H_Z$ and determine the $H_{int}(T)$ for different values of P. The $2\nu_Q$ transition at $P = 0$ shown in the upper panel of Fig.1 is asymmetrically split into two resonances by H_{int}, consistently with the previous result.[6] By contrast, the resonance frequency ν_p of the $3\nu_Q$ transition is decreased by H_{int} as seen in Fig.1. The T dependence of ν_p at P=0, 0.46, 1.00 and 1.23 GPa is shown in Fig.2(a). T_N is marked by arrows in Fig.2(a) and is precisely determined as the temperature below which ν_p decreases. It is notable that T_N slightly increases from 3.8 K at P=0 to $\sim$ 4 K at $P = 1.00$ GPa, but decreases to $\sim$ 3.6 K at P=1.23 GPa. The occurrence of the magnetic ordering at $P = 1.23$ GPa is clearly corroborated by a distinct peak in $1/T_1T$ at $T_N = 3.6$ K that probes critical magnetic fluctuations toward the magnetic ordering as shown in the inset of Fig.2(a).

$H_{int} = 1.75$ kOe at $P = 0$ and $T = 1.4$ K is estimated from the size of ν_p reduction of the $3\nu_Q$ transition as well as the splitting of the $2\nu_Q$ transition. This is consistent with the previous result.[6] Note that ν_p is only sensitive to the magnitude of H_{int}. $H_{int}(P)$ is plotted against a reduced temperature $t = T/T_N$ in Fig.2(b). Unexpectedly, the saturated value of $H_{int} \sim 0.39$ kOe at $P = 1.23$ GPa is about five times smaller than $H_{int} \sim 1.75$ kOe at $P = 0$, although T_N changes moderately. This slight pressure dependence of T_N contrasts with the strong reduction of H_{int}. A recent neutron experiment reported that Ce-ordered moments ($M_Q = 0.264(4)\mu_B$ at 1.4 K and $P = 0$) that lie in the basal plane are antiferromagnetically aligned, but they spiral transversely along the c axis with an incommensurate wave vector $\mathbf{q}_{\rm M}$=(1/2, 1/2, 0.297).[7] $H_{int}(P)$ is then extrapolated to zero at $P^* = 1.6 \pm 0.1$ GPa (see Fig.4(a)). If $M_Q(P)$ is directed in the basal plane, the M_Q would be scaled to H_{int} and substantially reduced to $\sim 0.05\mu_B$ at $P = 1.23$GPa. On the other hand, if $M_Q(P)$ is rotated with P from the ab plane to the c axis, the angle θ between the direction of M_Q and the c axis would be progressively smaller, extrapolated to zero at $P^* = 1.6 \pm 0.1$ GPa (see Fig.4(a)). This is because H_{int} at the In(1) site is canceled out at θ=0. H_{int} orignates from the direct dipolar field from the Ce ordered moments that reaches 30% of the total and from the indirect "pseudo" dipolar (anisotropic) field via the supertransferred hyperfine interaction. The latter internal field acts on the In site through the hybridization between In $5p$- and Ce $4f$-orbits. Note that the isotropic hyperfine field originating from Ce ordered moments is canceled out at the In(1) site. In order to see which P-induced change is more likely in the AF state, we need to consider the T dependence of $H_{int}(t)/H_{int}(0)$ displayed in the inset of Fig.2(b), where $H_{int}(0)$ is the low-T saturated value. A rapid growing of $H_{int}(T)$ is evident even at P=1.23 GPa. It would be therefore unlikely that some itinerant magnetic ordering takes place with a *reduced moment* and rather likely that the ordered moments rotate toward the c axis. As a result, it might be expected that the spiral order evolves into some commensurate AF fluctuation regime at $P^* = 1.6 \pm 0.1$ GPa. Note that P^* is close in value to a critical pressure $P_c \sim 1.63$ GPa which was suggested from the resistivity measurement.[5] To resolve this issue, further neutron experiment under pressure is highly desired.

We next deal with the SC region. The T_1 in the SC and normal state at $P = 2.1$ GPa was measured at the $1\nu_Q$ and $2\nu_Q$ transitions in order to avoid heating effect due to rf-excitation pulses. T_1 was determined by a single component. Figure 3 shows the T dependence of $^{115}(1/T_1)$ at $P = 0$ and 2.1 GPa. $^{115}(1/T_1)$ exhibits no coherence peak just below T_c=2.2 K, followed by a T^3 dependence down to $\sim$ 0.3 K. This is a convincing experimental evidence for the unconventional nature of the P-induced superconductivity in $CeRhIn_5$. Likewise all previous examples, a line-node gap model is applicable to the SC state in $CeRhIn_5$. Assuming an anisotropic energy gap model with $\Delta = \Delta_0 \cos\theta$, a solid line in Fig.3 is a fit for the $^{115}(1/T_1)$ data with $2\Delta_0 = 5k_{\rm B}T_c$.

We argue magnetic characters in the normal state at P =2.1 GPa. According to the SCR theory for nearly-AF metals by Moriya *et al*,[8] $1/T_1T \propto \chi_Q(T)^n$. Here a power-law dependence of the staggered susceptibility $\chi_Q(T)$ is obtained as n=1 and 1/2 for two (2D) and three (3D) dimensional electronic systems, respectively. By noting that $\chi_Q(T)$ follows a Curie-Weiss law of $1/(T+\theta)$, a behavior of $(T_1T)^2 \propto (T+\theta)$ is expected for the 3D nearly AF regime. As a matter of fact, as indicated in the inset of Fig.3, a fit of $(T_1T)^2 \propto (T+\theta)$ with $\theta = 1.5$ K is consistent with the present result in a relatively wide T range of T_c = 2.2 K – 30 K. This shows that 3D AF fluctuations are dominant in the normal state at P=2.1GPa.[8, 9, 10].

Hegger *et al.* speculated that the maximum at $T_{\chi m}$ =7.5 K and $P = 0$ in the susceptibility is associated with the development of 2D AF correlations in the $CeIn_3$ layers,[5] since a 2D-like magnetic character is expected from its quasi-2D crystal structure in the lower P region. With increasing P, $T_{\chi m}$ decreases approximately linearly and it would be extrapolated to $T = 0$ at $P_m = 1.3 \pm 0.4$ GPa. This is indicative of the 2D character in magnetic properties, which may be progressively suppressed as P increases. It is noteworthy that P_m is comparable to $P_c \sim 1.63$ GPa and $P^* = 1.6 \pm 0.1$ GPa. Therefore, at $P = 2.1$GPa exceeding either P_m or P^*, it is considered that AF fluctuations possess a 3D character rather than a 2D one. However, further works are needed to elucidate the role of critical AF fluctuations in the onset of the unconventional P-induced superconductivity in $CeRhIn_5$.

Figure 4(a) presents a phase diagram of the AF and SC phases along with the previous results.[5] According to the Ref.[4], at P_c ~1.63 GPa, the SC transition in the resistivity measurement begins around 2 K and reaches a zero-resistance state with a broad transition width.[5] In agreement, as shown in Fig.4(b), we found that the onset temperature in ac-χ is in accord with this zero-resistance T_c. However the size of the SC diamagnetism at 1.4 K in ac-χ is substantially reduced. At $P = 1.5$ GPa, no change in ac-χ is observable at all, supporting a critical pressure P_c ~1.63 GPa as suggested in the previous work.[5] Therefore it was ensured from the present ac-χ measurement that the bulk SC transition takes place down to $P = 1.8$ GPa, but probably not at pressures lower than $P = 1.65$ GPa. The present NQR study confirms that T_N slightly increases up to $T_N \sim 4$ K at P=1.00 GPa, but decreases to 3.6 K at P=1.23 GPa. The internal field H_{int} is extrapolated to zero at $P^* = 1.6 \pm 0.1$ GPa which is close to P_c ~1.63 GPa. Either the reduction in the ordered moment M_Q or its rotation from the ab plane to the c axis may occur as P increases. From the rapid growing of $H_{int}(P)$ upon cooling below T_N, we believe that the rotation of $M_Q(P)$ occurs with P, but its marked reduction does not. In this context, we suggest that the spiral order is presumably suppressed across a critical pressure $P^* = 1.6 \pm 0.1$ GPa. Eventually, the SC transition emerges in $CeRhIn_5$ at pressures exceeding P^*. It is highly desired to elucidate whether AF or SC fluctuations prevent the onset of any type of long-range orders in the vicinity of $P^* = 1.6 \pm 0.1$ GPa.

In conclusion, we have reported that unconventional magnetic and superconducting states are induced by applying P to the HF-AF $CeRhIn_5$. In the magnetic region, T_N exhibits a moderate variation. By contrast, H_{int} whose presence is due to the magnetic ordering is unexpectedly reduced at $P = 1.23$ GPa, extrapolated to zero at $P^* = 1.6 \pm 0.1$GPa. The spiral order might be suppressed presumably due to the rotation of the ordered moments toward the c axis. This P^* is comparable to P_c ~1.63 GPa at which the bulk SC transition is not emergent as suggested from the previous resistivity [5] and corroborated by the present ac-χ measurements. In the SC state at $P = 2.1$GPa, we found $1/T_1 \propto T^3$ that shows the existence of line-nodes in the gap function. In the normal state, the remarkable behavior of $(1/T_1T) \propto 1/\sqrt{T + 1.5}$ which is consistent with the 3D nearly AF fluctuation regime suggests that the magnetic nature possesses a 3D-like character at pressures where the bulk SC sets in. This work was supported by the COE Research (10CE2004) in Grant-in-Aid for Scientific Research from the Ministry of Education, Sport, Science and Culture of Japan. One of the authors (T.M.) has been supported by JSPS Research Fellowships for Young Scientists.

[1] D. Jaccard, K. Behnia, and J. Sierro, Phys. Lett. A **163**, 475 (1992).
[2] F. M. Grosche, S. R. Julian, N. D. Mathur, and G. G. Lonzarich, Physica B **223-224**, 50 (1996).
[3] N. D. Mathur, F. M. Grosche, S. R. Julian, I. R. Walker, D. M. Freye, R. K. W. Haselwimmer, and G. G. Lonzarich, Nature **394**, 39 (1998).
[4] See *e.g.*, Y. Kitaoka, H. Tou, G. -q. Zheng, K. Ishida, K. Asayama, T. C. Kobayashi, A. Kohda, N. Takeshita, K. Amaya, Y. Onuki, G. Geibel, C. Schank and F. Steglich, Physica B **206 & 207**, 55 (1995); K. Ishida, Y. Tokunaga, Y. Kitaoka, G. -q, Zheng, K. Magishi, H. Mukuda, H. Tou, T. Mito and K. Asayama, Physica B **259-261**, 511 (1999).
[5] H. Hegger, C. Petrovic, E. G. Moshopoulou, M. F. Hundley, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Phys. Rev. Lett. **84**, 4986 (2000).
[6] N. J. Curro, P. C. Hammel, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, and Z. Fisk, Phys. Rev. B **62**, R6100 (2000).
[7] Wei Bao, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, Z. Fisk, J. W. Lynn, and R. W. Erwin, Phys. Rev. B **62**, R14 621 (2000).
[8] T. Moriya and T. Takimoto, J. Phys. Soc. Jpn. **64**, 960 (1995).
[9] A. Ishigaki and T. Moriya, J. Phys. Soc. Jpn. **65**, 3402 (1996).
[10] S. Nakamura, T. Moriya, and K. Ueda, J. Phys. Soc. Jpn. **65**, 4026 (1996).

FIG. 1: ^{115}In-NQR spectra at various values of pressure. The upper panel indicates the $2\nu_Q$ and $3\nu_Q$ transitions below T_N. The lower panel indicates the ^{115}In-NQR spectrum at ambient pressure ($P = 0$) above T_N. In the absence of internal field, the In(1) spectrum consists of four transitions given by $\nu = n\nu_Q$, where n =1, 2, 3 and 4 (see text). Below T_N, the $2\nu_Q$ and $3\nu_Q$ transition splits asymmetrically and shifts, respectively.

FIG. 2: (a) Temperature dependence of the resonance frequency ν_p of the $3\nu_Q$ transition at P=0, 0.46, 1.00 and 1.23 GPa. The arrow indicates T_N. The inset shows the temperature dependence of $^{115}(1/T_1T)$ at P=1.23GPa. (b) Temperature dependence of the internal field H_{int} is plotted against $t = T/T_N$ at P=0, 0.46, 1.00 and 1.23 GPa. The inset indicates $H_{int}(t)/H_{int}(0)$ vs t plots. Here $H_{int}(0)$ is a saturated value at low temperature.

FIG. 3: Temperature dependence of ^{115}In nuclear spin-lattice relaxation rate, $^{115}(1/T_1)$ at $P = 2.1$ GPa along with the data at $P = 0$ both displayed in logarithmic scales. The solid line is a fit assuming a line-node gap $\Delta(\phi) = \Delta_0 \cos\phi$ with $2\Delta_0 = 5k_{\rm B}T_c$. Inset: $(T_1T)^2$ vs T plot at $P = 2.1$ GPa. The solid line is a fit based on the 3D-SCR theory [8, 9, 10] that predicts the following behavior: $(T_1T)^2 \propto 1/\chi_Q(T) \propto (T+\theta)$ where $\theta = 1.5$ K. Note that $\chi_Q \propto (T+\theta)^{-1}$ follows a Curie-Weiss law for the nearly AF Fermi-liquid regime.

FIG. 4: (a) The pressure dependences of T_N (open triangles), T_c (open circles) and H_{int} (open diamonds) determined from the present work are shown together with the previous data.[5] The $H_{int}(P)$ is extrapolated to zero at $P^* = 1.6 \pm 0.1$GPa as indicated by the dotted line. If the reduction of $H_{int}(P)$ is attributed to the rotation of $M_Q(P)$, $H_{int}(P)$ is proportional to sinθ (See text). The SC transition width was marked by bars: $T_{\rm c}^{\rm onset} - T_{\rm c}^{\rm offset}$ defined in Fig.4(b). The solid lines are guides to the eye.
(b) Temperature dependence of the high-frequency ac susceptibility (ac-χ) measured using an *in-situ* NQR coil at various values of pressure. $T_{\rm c}$ is defined as the temperature at which the ac-χ decreases to 10% of the total Meissner signal at each pressure. $T_{\rm c}^{\rm onset}$ and $T_{\rm c}^{\rm offset}$ are defined as the respective temperature at which the SC diamagnetism starts to emerge and the ac-χ reaches to 90% of the total Meissner signal.

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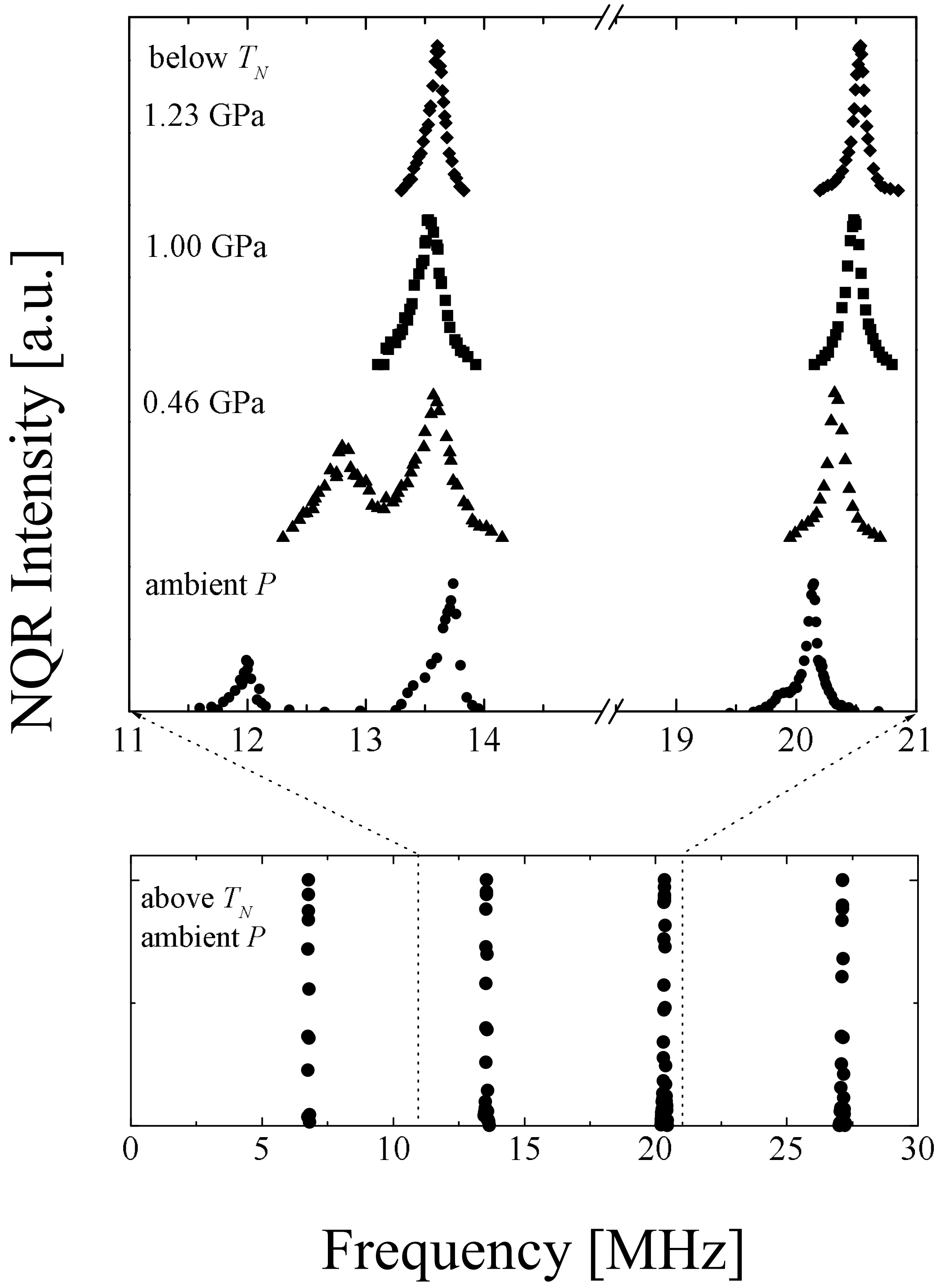

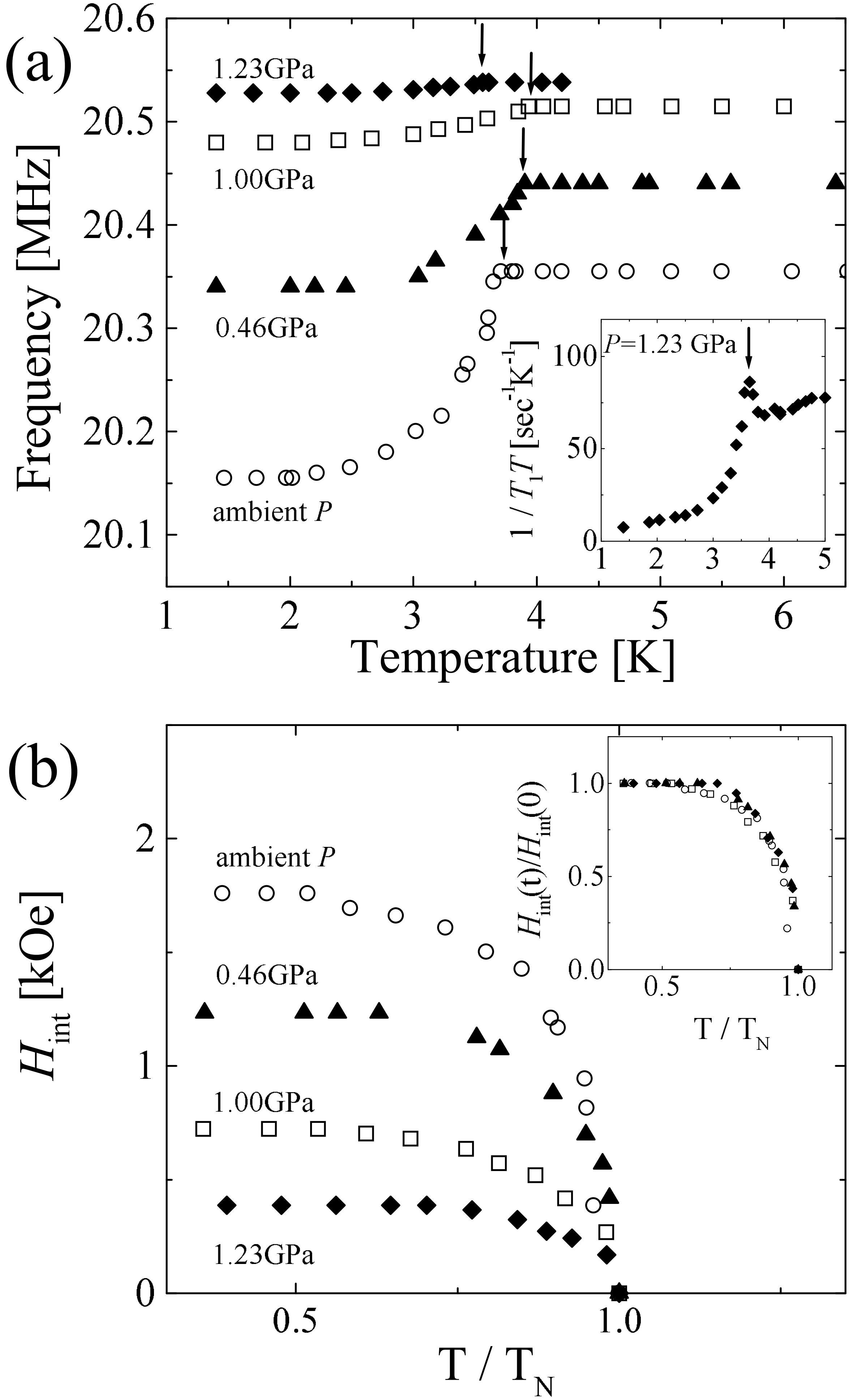

(a)
Frequency [MHz]
Temperature [K]
1.23GPa
1.00GPa
0.46GPa
ambient P
20.6
20.5
20.4
20.3
20.2
20.1
1
2
3
4
5
6
P=1.23 GPa
1 / T1T [sec-1K-1]
100
50
0
(b)
Hint [kOe]
T / TN
ambient P
0.46GPa
1.00GPa
1.23GPa
2
1
0
0.5
1.0
Hint(t)/Hint(0)

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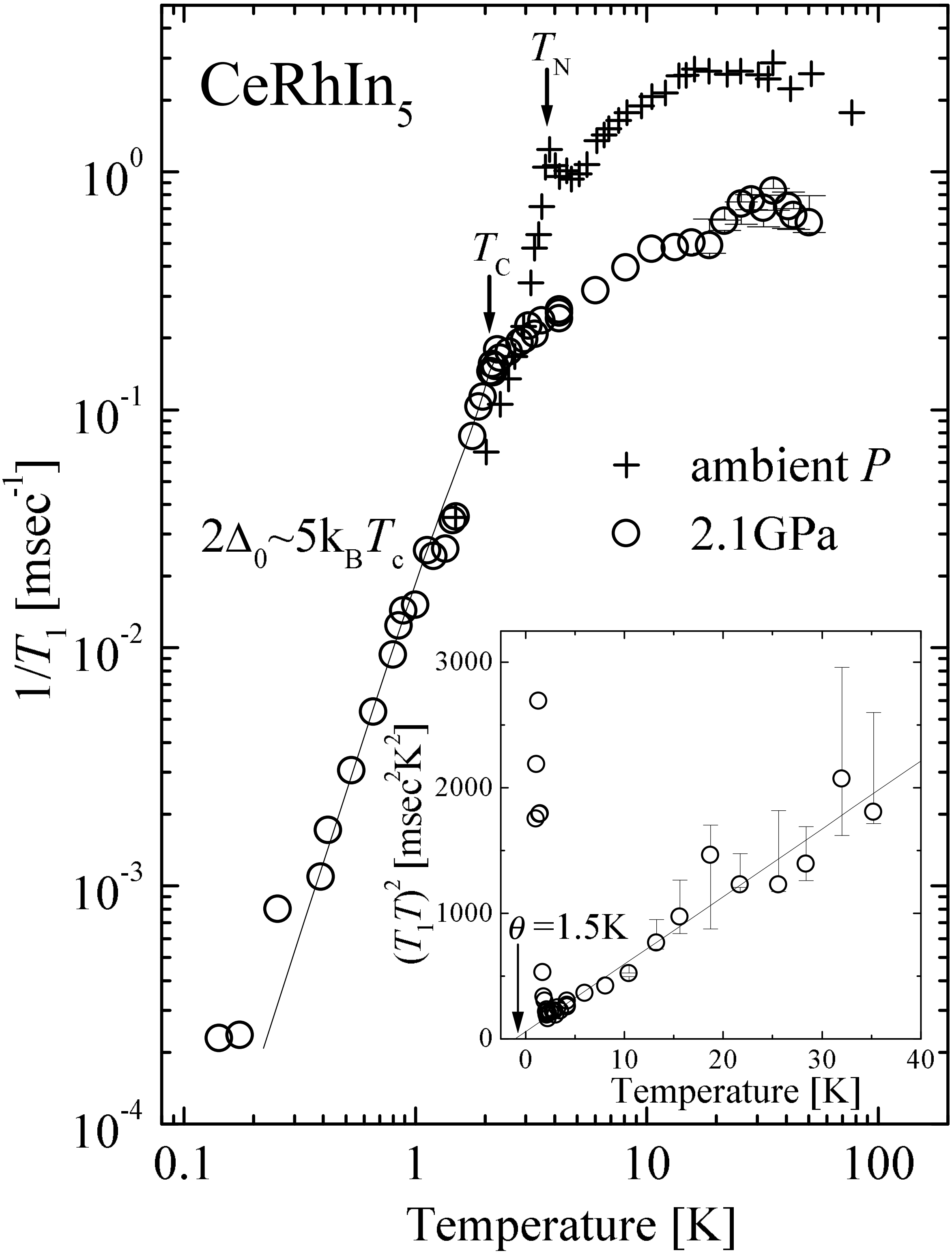
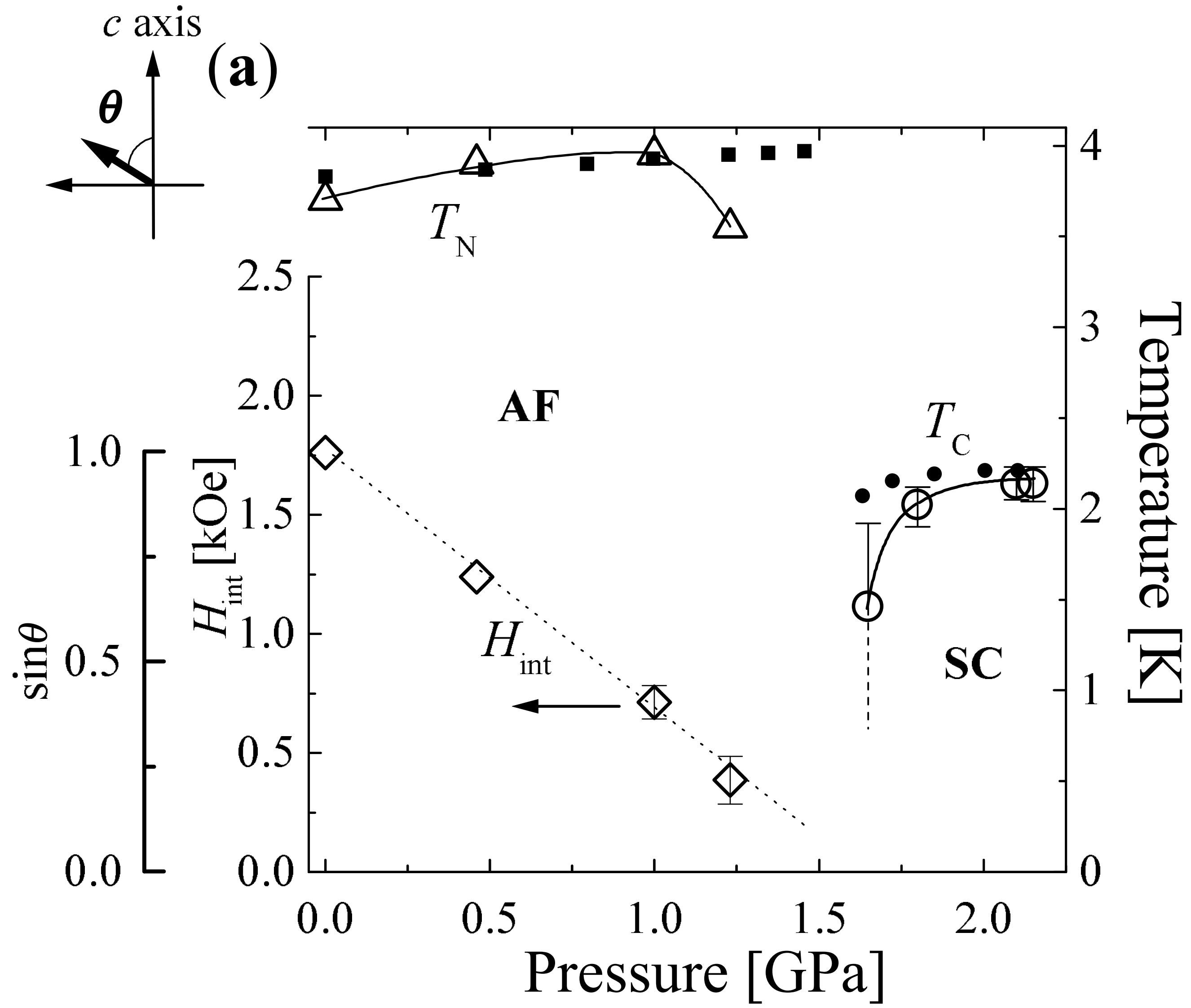
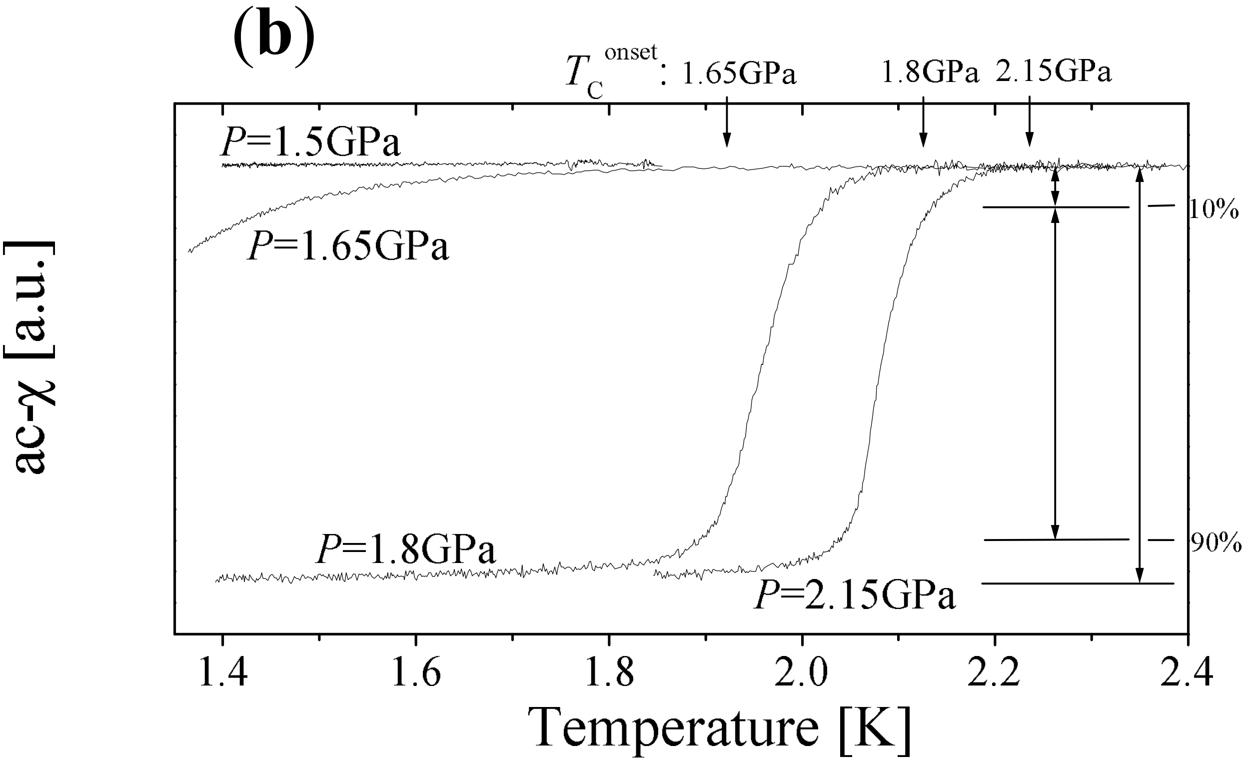




c axis
(a)
θ
T_N
AF
T_C
SC
H_int
sinθ
H_int [kOe]
Temperature [K]
Pressure [GPa]



(b)
T_C^onset: 1.65GPa
1.8GPa
2.15GPa
P=1.5GPa
P=1.65GPa
P=1.8GPa
P=2.15GPa
10%
90%
ac-χ [a.u.]
Temperature [K]