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^{59}Co NMR shift anomalies and spin dynamics in the normal state of superconducting $CeCoIn_5$: Verification of two-dimensional antiferromagnetic spin fluctuations

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We have measured the Knight shifts (K) and nuclear relaxation times (T_1) for ^{59}Co in $CeCoIn_5$ under external fields along a and c axes with the goal of establishing the anisotropy of antiferromagnetic (AFM) spin fluctuations (SF). In our approach, we revisit the problem of interpreting anomalies in the relationship between Knight shift $K_{a,c}$ and static susceptibility $\chi_{a,c}$: assuming a single component susceptibility implies a temperature-dependent hyperfine coupling $A_{a,c}(T)$. Once adopted, a known discrepancy between the behaviors of T_1^{-1} for ^{59}Co and ^{115}In(1) sites is eliminated to within experimental uncertainties and the variation with temperature is analyzed within the framework of two-dimensional AFM SF in proximity to a quantum-critical point. Moreover, the ratio $[T_{1a}/T_{1c}]$ indicates easy-plane anisotropy for $T<T^* \sim 40$ K, for the heavy-fermion state.

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The need to determine the nature of spin fluctuations and their possible relationship to unconventional superconductivity in heavy-fermion (HF) compounds, and strongly correlated systems more broadly, has posed a long-standing problem. The HF superconductors $CeCoIn_5$ (T_c=2.3 K) (Ref. 1) and $PuCoGa_5$ (T_c=18.5 K) (Ref. 2) provide interesting case studies for this problem. Both compounds can be classified as members of a "115" family with the layered $HoCoGa_5$-type crystal structure shown in the inset of Fig. 1(a). Systematic NMR investigations of the Ce115 systems have established that these superconductors have d-wave superconducting gaps,[3–6] and that antiferromagnetic (AFM) spin fluctuations (SF) play an active role in the superconducting pairing.[7–10] NMR measurements have similarly demonstrated evidence for d-wave superconductivity in the Pu115 systems[11,12] while T_c values are an order of magnitude higher than those for the Ce115 systems. Recently, Kambe *et al.*[13] has proposed from K-T_1 analyses in the 115 family that there may be a favorable magnetic anisotropy for d-wave superconductivity in f-electron systems, namely, AFM XY-type magnetic anisotropy, rather than Heisenberg or Ising type. Moreover, such an AFM XY-type anisotropy in $PuRhGa_5$ with T_c=8.5 K is found to be strong, in clear contrast to other nonsuperconducting An115 (An=U, Np) systems. In order to broaden an examination of magnetic anisotropy to Ce115 from An115 systems, we report in this Rapid Communication on the magnetic anisotropy in $CeCoIn_5$ from K-T_1 analyses using ^{59}Co NMR and compare to results obtained in $PuRhGa_5$.

The anisotropy of AF spin fluctuations is inferred from spin lattice relaxation measurements T_1^{-1} of ^{59}Co nuclear spins. T_1^{-1} is written

$$\frac{1}{T_1} = \frac{k_B T}{(\gamma_e \hbar)^2} 2(\gamma_n A_\perp)^2 \sum_{\boldsymbol{q}} f_\perp^2(\boldsymbol{q}) \frac{\mathrm{Im}\, \chi_\perp(\boldsymbol{q},\omega_0)}{\omega_0}, \quad (1)$$

where γ_n and γ_e are the nuclear and electronic gyromagnetic ratios, A is the transferred hyperfine coupling constant, $f_\alpha(\boldsymbol{q})$ is the hyperfine form factor, Im $\chi(\boldsymbol{q},\omega_0)$ is the imaginary part of dynamical susceptibility, ω_0 is the nuclear Larmor frequency, and the suffix $\perp$ refers to the component perpendicular to the quantization axis. The observed anisotropic hyperfine coupling constants mainly originate from the hy-

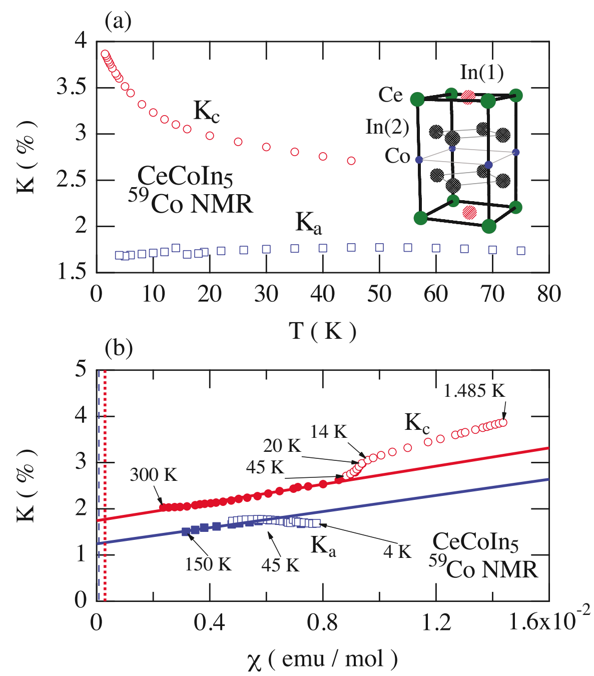

FIG. 1. (Color online) (a) Temperature dependence of Knight shifts for ^{59}Co in the normal state of $CeCoIn_5$ under the external field of 50 kOe along a and c axes. The inset shows the crystal structure of $CeCoIn_5$. (b) $K_{a,c}$-χ plot for ^{59}Co in $CeCoIn_5$. The closed circles and squares are reproduced from Ref. 3. The solid lines represent the K-χ lines determined in the temperature range from 300 to 40 K. The dashed and dotted lines mean the temperature-independent term (χ_0) of static susceptibility along a and c axes, respectively.

bridization between Ce $4f$ and the ligand $5p/3d$. Therefore, the $\boldsymbol{q}$ dependence of the transferred hyperfine coupling is imposed by $f_\alpha(\boldsymbol{q})$ since the transferred hyperfine fields are locally produced from nearest Ce ions.

Our analysis of the SF contribution to ${}^{59}T_1^{-1}$ is based on the assumption that the dynamic susceptibility is comprised of a single component, meaning that Eq. (1) applies. Consequently, the (temperature-dependent) hyperfine coupling is inferred directly from the Knight shift results shown in Fig. 1 and the relationship $K_{a,c}=A_{a,c}\chi_{a,c}+K_0$ with K_0 independent of temperature. The data were obtained using sufficiently small rf power so as to eliminate heating from eddy currents and are similar to those reported in Ref. 3. We will return to address the validity of the single-component response later. In particular, we contrast these implications relative to a susceptibility consisting of two components: a contribution from local moments dominant at high temperatures and a contribution from the heavy-fermion part dominating at low temperatures.[14,15]

The temperature-dependent slope appearing in Fig. 1 is attributed to a variation in the hyperfine interaction from a single-component dynamical susceptibility. For high temperatures, $A(T)$ is defined as usual, and $A(T\lesssim 40\ \mathrm{K})$ is defined by the relation $nA(T)=[K(T)-K_0]/[\chi(T)-\chi_0]$, where n is the number of the nearest-neighbor Ce atoms, χ_0 the temperature-independent part of the static susceptibility, determined from the Curie-Weiss fit $\chi(T)=\chi_0+C/(T-\Theta)$ applied to the high-T region, with C the Curie constant, and Θ the Weiss temperature. A similar procedure was previously applied to the U115 systems.[16] The derived $A(T)$ is plotted in Fig. 2(a). As shown in Fig. 2, this transferred $A(T)$ for ^{59}Co for both directions varies gradually for $T\lesssim 40$ K and saturates for $T\lesssim 10$ K. A similar procedure applied to the In(1) site, using the data reported in Ref. 5, leads to the results shown in Fig. 2(b). Interestingly, the hyperfine coupling constant along the a axis $[{}^{115}A_a(1)]$ for the In(1) sites has no significant temperature dependence. It is noted that the reported data for In(1) site are substantially different from that by the other authors.[3] However, these data in Ref. 3 also provide a T-independent coupling constant for In(1) in the case of $H_0\|a$. $[{}^{115}A_c(1)]$ decreases smoothly upon lowering the temperature. In what follows, these results are used to interpret the ^{59}Co $T_1^{-1}(T)$ and establish the anisotropy of the AFM spin fluctuations.

The temperature dependence of the ${}^{59}T_1^{-1}$ is shown in inset of Fig. 3 for magnetic field directions $H\|(a,c)$, and the In(1) results from nuclear quadrupole resonance (NQR) are reproduced from Ref. 10. For the latter case, the observation of ${}^{115}T_1^{-1}\sim T^{1/4}$ is considered as an evidence for proximity to an AFM quantum-critical point (QCP). Since the quantization axis is c axis for both ^{59}Co NMR $(H\|c)$ and ^{115}In(1)-NQR, which corresponds to in-plane SF by Eq. (1), the steeper power law for ^{59}Co, ${}^{59}T_1^{-1}\sim T^{1/2}$ over a comparable temperature range, is puzzling. To emphasize the behavior of the dynamical susceptibility, we normalize the relaxation rates for $H\|c$ by $[{}^{59}A_a(T)]^2$, and show the result in the main panel of Fig. 3. For In(1) sites, the normalization simply rescales the data of the inset because the temperature dependence of ${}^{115}A_a(T)$ is negligible. The effect of the normalization on the ^{59}Co data is significant: by taking the result shown in Fig. 1

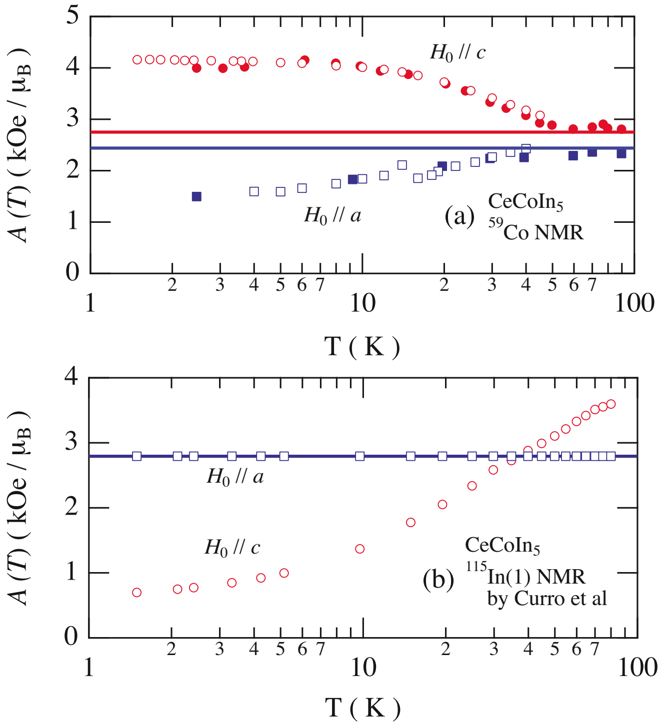

FIG. 2. (Color online) Temperature dependence of transferred hyperfine coupling constants for (a) ^{59}Co and (b) ^{115}In(1) in $\mathrm{CeCoIn_5}$ along a and c axes. The In(1) sites means the tetragonal In sites in the Ce-In plane, of which data are derived from the data by Curro *et al.* (Ref. 5). The solid lines represent the hyperfine coupling constants in the high-temperature range.

to mean that ${}^{59}A={}^{59}A(T)$, then the dynamical susceptibility in Eq. (1) inferred from both sites has a unique temperature dependence $(\propto T^{1/4})$ and motivates our consideration of a single-component model. For completeness, we note that the Korringa behavior of the normalized $T_1^{-1}\sim T$ for $T\lesssim 1$ K in the case of $H\|c$ is associated with a crossover from quantum-critical to Fermi-liquid behavior, consistent with conclusions of electrical resistivity measurements.[18]

Having proposed that the K-χ anomalies result from variations in the hyperfine interaction, the analysis of the nuclear spin relaxation proceeds similarly to that described in Ref. 13. The rate $R_\alpha\equiv k(\gamma_n A_\alpha)^2\Sigma_{\boldsymbol{q}}\mathrm{Im}\,\chi_\alpha(\boldsymbol{q},\omega_0)/\omega_0$ $(\alpha=a,c)$ is defined where k is a numerical constant of $k_B/(\gamma_e\hbar)^2$. $f_\alpha^2(\boldsymbol{q})$ is ignored for the Co site, and from Eq. (1),[19] $(1/T_1T)_{H_0\|c}=2R_a$ and $(1/T_1T)_{H_0\|a}=R_a+R_c$. Taking a Lorentzian form for $\mathrm{Im}\,\chi_\alpha(\boldsymbol{q},\omega_0)$ with the magnetic fluctuation energy $\Gamma(\boldsymbol{q})$, $\mathrm{Im}\,\chi_\alpha(\boldsymbol{q},\omega_0)/\omega_0=\chi_\alpha(\boldsymbol{q})/\Gamma_\alpha(\boldsymbol{q})$. Then, within the strongly correlated limit approximation $[2\pi\chi_\alpha(q)\Gamma_\alpha(q)\sim 1]$,[20] the q-averaged (local) magnetic fluctuation energy $\Gamma_\alpha\equiv[\overline{\Gamma_\alpha(\boldsymbol{q})^2}]^{1/2}$ is given as $\Gamma_\alpha=[k\gamma_n^2A^2(T)/2\pi R_\alpha]^{1/2}$. The obtained Γ_α is plotted in the inset of Fig. 4. For comparison, the similar quantity obtained from inelastic neutron scattering (INS) at $Q=(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ (Ref. 21) is also plotted in the inset of Fig. 4, which corresponds to the perpendicular component to Q. The magnitude of Γ_α obtained is consistent with the INS result in the normal state, as well as the In(1)-NMR (Ref. 7) although the INS results indicate easy-axis fluctuations near the superconduct-

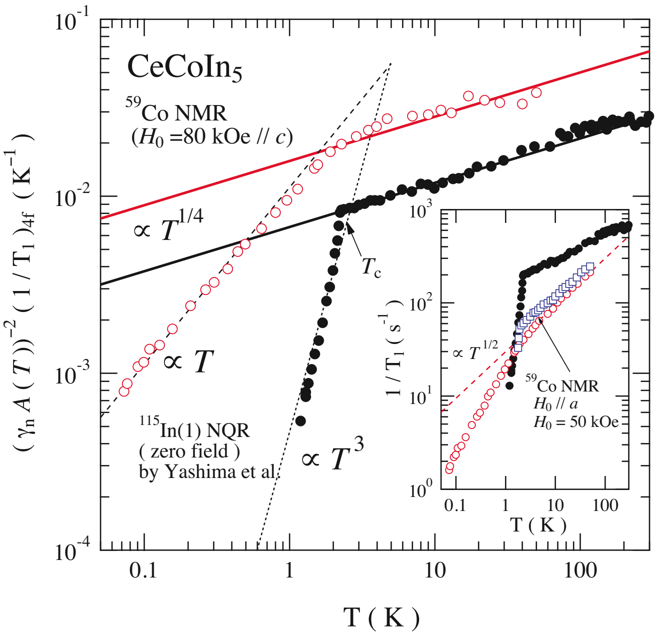

FIG. 3. (Color online) The temperature dependence of T_1^{-1} normalized by the square of hyperfine coupling constant $[\gamma_n A_a(T)]^2$ for ^{59}Co NMR (○) and ^{115}In NQR (●) after subtractions of those in $LaCoIn_5$ (Ref. 17). The latter are reproduced from Ref. 10. Measurements for $H \| c$ are made in a field well above $H_{c2}(0)=49.5$ kOe. The inset shows the raw T_1^{-1} for ^{59}Co NMR along a (□) and c (○) axis, and for ^{115}In(1) NQR (Ref. 10) as well.

ing state. It is also suggested by Curro *et al.*[7,22] that the Γ_α in the high-T region above ~ 50 K is quite similar to that of the antiferromagnet $CeRhIn_5$, which orders at $T_N=3.8$ K in an incommensurate magnetic structure.

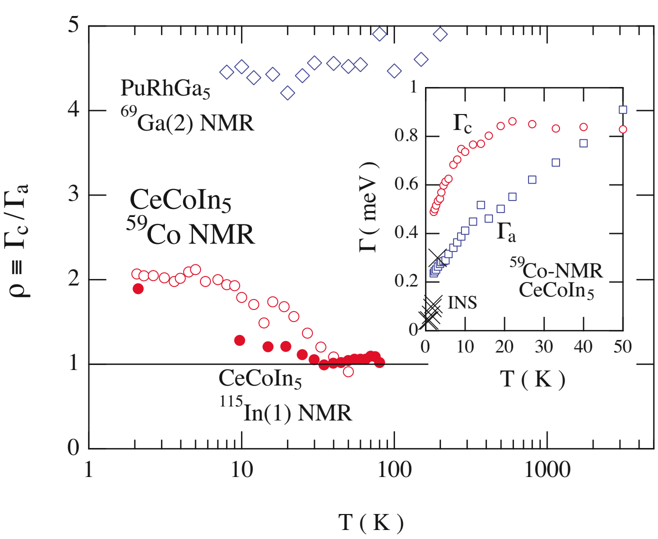

FIG. 4. (Color online) Temperature dependence of the local magnetic fluctuation energy ratio $\rho \equiv \Gamma_c/\Gamma_a$ for $CeCoIn_5$, which are computed from the ^{59}Co NMR data (○) and ^{115}In(1) NMR (Ref. 22) (●) and from ^{69}Ga(2) for $PuRhGa_5$ (◇). The inset shows the temperature dependence of $\Gamma_{a,c}$ for $CeCoIn_5$. The crosses (×) in the inset mean the data obtained by inelastic neutron scattering at $Q=(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ (Ref. 21).

A notable feature in the Γ_α-T plot is the anisotropy between Γ_a and Γ_c. Shown in Fig. 4 is an anisotropy factor $\rho \equiv \Gamma_c/\Gamma_a$ that increases gradually below $T^* \sim 40$ K. It changes from the isotropic value $\rho \sim 1$ to ~ 2 around 1 K. $\rho > 1$ indicates XY-type magnetic anisotropy. For comparison, the same quantity ρ for the d-wave superconductor $PuRhGa_5$ is plotted in Fig. 4, which is updated from the previous data.[13] Although the T dependence of ρ is weak for $PuRhGa_5$, it is remarkably large as compared with $CeCoIn_5$ and the other actinide 115 compounds where ρ typically is $\lesssim 1$. It is an open question whether a strongly AFM XY type is more favorable than a nearly isotropic one for d-wave HF superconductors and further experiments in 115 systems will strengthen the hypothesis of an association.

We return now to discuss generally the interpretation of the Knight shift anomaly in heavy-fermion materials. A large number of heavy-fermion systems have exhibited nonlinear relationships in K-χ similar to what is shown in Fig. 1, and a number of explanations have been proposed. For example, as in this work, the anomaly has been attributed to a temperature-dependent hyperfine coupling. Then, the change in $A(T)$ should, in some way, be related to the emergence of the heavy-fermion state. Alternatively, the universality of a *collective* mechanism linking the anomaly to the onset of the formation of the heavy electron liquid, makes for a particularly interesting proposal.[14] Phenomenological support for the two-fluid interpretation is presented in Ref. 15, in which Ce-, Yb-, and U-based systems are shown using a scaling argument to exhibit a heavy-fermion susceptibility growing as log T below a coherence temperature T^*. In the context of the hyperfine fields, the analysis involves two independent terms appearing on the right-hand side of Eq. (1), one for the coupling to localized f moments, and the other for the coupling to the heavy-fermion component. With regard to $CeCoIn_5$, it was previously shown that the In(1) site is insensitive to T^*.[5] As a careful balancing of parameters is required to produce this result, these circumstances also motivated the approach here. Also, the two fluid approach generically gives a different background shift (K_0) for $T > T^*$ and $T < T^*$, which means that the orbital Knight shift would escalate abruptly at T^*. Especially it is anomalously large for K_a in Fig. 1(b) since the χ_0 does not change significantly through T^*.

On the other hand, it is not straightforward to suggest a mechanism for a temperature-dependent hyperfine coupling in $CeCoIn_5$. For example, it is unlikely that such a change in coupling constant results from thermal crystal field level occupations because the anomalous Knight shift behavior occurs too abruptly. Perhaps the change in the hyperfine couplings may be related to the increase in anisotropic f-p hybridization microscopically proposed by dynamical mean field theory (DMFT) calculations in combination with local-density approximation.[23] In addition, very recently, the similar analysis based on In(1)- and In(2)-NMR results for $CeIrIn_5$ is successful to explain the coherent fashion in heavy-fermion system.[24] Within the context of $CeCoIn_5$, the analysis shown here provides a self-consistent framework in which to view the temperature dependence of the properties of the hyperfine fields without the need for introducing a two-component susceptibility.

In conclusion, we have motivated a reinterpretation of the nonlinear K-χ relationship observed in the 115 materials. Specifically, we consider the implications for attributing the anomalous behavior entirely to a temperature-dependent hyperfine interaction ${}^{59}A = {}^{59}A(T)$. As measured, $1/T_1$ for ^{59}Co NMR in $CeCoIn_5$ shows a $T^{1/2}$ behavior under external fields along c axis in the high-T region. Meanwhile, $1/T_1$ for ^{115}In(1) NQR shows a $T^{1/4}$ behavior in the normal state under zero field, which is consistent with strong enhancement of two-dimensional (2D) AFM SF near QCP. This apparent discrepancy of T exponent of $1/T_1$ on the different sites in the same material cannot be reconciled using T-independent hyperfine coupling constants. However, normalizing the rates by the square of the derived $A(T)$ leads to a similar $T^{1/4}$ dependence even for ^{59}Co NMR in the high-T region, which suggests the unique component of Im $\chi(\boldsymbol{q},\omega_0)$ in $CeCoIn_5$ and the robustness of 2D AFM nature even under a magnetic field along the c axis. The AFM XY-type magnetic anisotropy increases below T^* and saturates near $\rho \sim 2$, a ratio much smaller than in the relatively higher T_c superconductor $PuRhGa_5$. Additional studies of the anisotropy and magnitude of spin fluctuations will be useful to establish a general framework for understanding their role in unconventional superconductors.

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[1] C. Petrovic, P. G. Pagliuso, M. F. Hundley, R. Movshovich, J. L. Sarrao, J. D. Thompson, Z. Fisk, and P. Monthoux, J. Phys.: Condens. Matter **13**, L337 (2001).

[2] J. L. Sarrao, L. A. Morales, J. D. Thompson, B. L. Scott, G. R. Stewart, F. Wastin, J. Rebizant, P. Boulet, E. Colineau, and G. H. Lander, Nature (London) **420**, 297 (2002).

[3] Y. Kohori, Y. Yamato, Y. Iwamoto, T. Kohara, E. D. Bauer, M. B. Maple, and J. L. Sarrao, Phys. Rev. B **64**, 134526 (2001).

[4] G.-q. Zheng, K. Tanabe, T. Mito, S. Kawasaki, Y. Kitaoka, D. Aoki, Y. Haga, and Y. Ōnuki, Phys. Rev. Lett. **86**, 4664 (2001).

[5] N. J. Curro, B. Simovic, P. C. Hammel, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, and G. B. Martins, Phys. Rev. B **64**, 180514(R) (2001).

[6] T. Mito, S. Kawasaki, G.-q. Zheng, Y. Kawasaki, K. Ishida, Y. Kitaoka, D. Aoki, Y. Haga, and Y. Ōnuki, Phys. Rev. B **63**, 220507(R) (2001).

[7] N. J. Curro, J. L. Sarrao, J. D. Thompson, P. G. Pagliuso, Š. Kos, A. Abanov, and D. Pines, Phys. Rev. Lett. **90**, 227202 (2003).

[8] S. Kawasaki, T. Mito, Y. Kawasaki, G.-q. Zheng, Y. Kitaoka, D. Aoki, Y. Haga, and Y. Ōnuki, Phys. Rev. Lett. **91**, 137001 (2003).

[9] S. Kawasaki, G.-q. Zheng, H. Kan, Y. Kitaoka, H. Shishido, and Y. Ōnuki, Phys. Rev. Lett. **94**, 037007 (2005).

[10] M. Yashima, S. Kawasaki, Y. Kawasaki, G.-q. Zheng, Y. Kitaoka, H. Shishido, R. Settai, Y. Haga, and Y. Ōnuki, J. Phys. Soc. Jpn. **73**, 2073 (2004).

[11] N. J. Curro, T. Caldwell, E. D. Bauer, L. A. Morales, M. J. Graf, Y. Bang, A. V. Balatsky, J. D. Thompson, and J. L. Sarrao, Nature (London) **434**, 622 (2005).

[12] H. Sakai *et al.*, J. Phys. Soc. Jpn. **74**, 1710 (2005).

[13] S. Kambe *et al.*, Phys. Rev. B **75**, 140509(R) (2007).

[14] S. Nakatsuji, D. Pines, and Z. Fisk, Phys. Rev. Lett. **92**, 016401 (2004).

[15] N. J. Curro, B. L. Young, J. Schmalian, and D. Pines, Phys. Rev. B **70**, 235117 (2004).

[16] H. Kato, H. Sakai, Y. Tokunaga, Y. Tokiwa, S. Ikeda, Y. Ōnuki, S. Kambe, and R. E. Walstedt, J. Phys. Soc. Jpn. **72**, 2357 (2003).

[17] Y. Kawasaki, S. Kawasaki, M. Yashima, T. Mito, G.-q. Zheng, Y. Kitaoka, H. Shishido, R. Settai, Y. Haga, and Y. Ōnuki, J. Phys. Soc. Jpn. **72**, 2308 (2003).

[18] J. Paglione, M. A. Tanatar, D. G. Hawthorn, E. Boaknin, R. W. Hill, F. Ronning, M. Sutherland, L. Taillefer, C. Petrovic, and P. C. Canfield, Phys. Rev. Lett. **91**, 246405 (2003).

[19] For the In(1) sites, $f^2(\boldsymbol{q}) = 16\cos^2(q_x a/2)\cos^2(q_y a/2)$, and for Co sites, $f^2(\boldsymbol{q}) = 4\cos^2(q_z c/2)$. Since SF in the paramagnetic (or normal) state usually have broad widths in $\boldsymbol{q}$ space, $1/T_1T$ can sense an AFM fluctuation beyond the moderate filtering by trigonometrical $f^2(\boldsymbol{q})$, except for the case of excessive *three* dimensional narrowing at a specific $\boldsymbol{q}$. Furthermore, the form factor has no anisotropy and should not play a role in the corresponding analysis of SF.

[20] T. Moriya and T. Takimoto, J. Phys. Soc. Jpn. **64**, 960 (1995).

[21] C. Stock, C. Broholm, J. Hudis, H. J. Kang, and C. Petrovic, Phys. Rev. Lett. **100**, 087001 (2008).

[22] N. J. Curro and D. Pines, J. Phys. Chem. Solids **68**, 2028 (2007).

[23] J. H. Shim, K. Haule, and G. Kotliar, Science **318**, 1615 (2007).

[24] S. Kambe, Y. Tokunaga, H. Sakai, H. Chudo, Y. Haga, T. D. Matsuda, and R. E. Walstedt, Phys. Rev. B **81**, 140405(R) (2010).