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One-component description of magnetic excitations in the heavy-fermion compound $CeIrIn_5$

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We report an ^{115}In NMR study of a single-crystal sample of the heavy-fermion compound $CeIrIn_5$. The observed nonlinear variation in Knight shift with static susceptibility is consistent with the two-fluid model of Nakatsuji *et al.* However, our results can also be understood in terms of a T-dependent hyperfine coupling, which accounts for the spin-lattice relaxation data naturally on the basis of a one-component dynamical susceptibility. In addition, the observed T dependence of the hyperfine coupling is scaled to a density of states given by dynamical mean-field theory.

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In heavy-fermion compounds, f electrons having a localized nature at high temperatures are considered to form heavy quasiparticles through coupling with the conduction electrons at low temperatures.[1] Although this crossover to a heavy-fermion state has not been explained in detail, a recently proposed two-fluid model describes the crossover phenomenologically on the assumption of separate localized and heavy-fermion components.[2,3] Further, it has been shown that the nonlinear relation between the NMR Knight shift K and the static susceptibility χ that is observed in many heavy-fermion compounds can be explained with a two-fluid model scaling law, which is characterized by a scaling temperature T^*.[4]

Considering the success of the two-fluid description of the static properties K and χ, it appears that two individual contributions to static susceptibility exist in heavy-fermion compounds. However, the nonlinear K-χ relation can also be explained in terms of a T-dependent hyperfine coupling. In the present study, such a variable coupling has been found, in addition, to be consistent with spin-lattice relaxation time T_1 results for ^{115}In in $CeIrIn_5$ in a picture based on a one-component dynamical susceptibility.

The heavy-fermion superconductor $CeIrIn_5$ has a large Sommerfeld coefficient $\gamma_{el}=0.75$ J/mol K^2 with a superconducting T_c of ~ 1.2 K.[5] High-quality single-crystal samples have been prepared by the Czochralski method.[6] The ^{115}In NMR ($I=9/2$, gyromagnetic ratio $\gamma_n=9.3295$ MHz/T), measurements were carried out using a phase-coherent pulsed spectrometer. The NMR spectra which we analyze are field-sweep spectra taken at constant frequency $\omega_n=59.8$ MHz. T_1 data were obtained using the standard spin-echo inversion-recovery method at $\omega_n \sim 75$ MHz.

In the $HoCoGa_5$($I/4mmm$)-structure compound $CeIrIn_5$, there are two crystallographically inequivalent In sites [In(1): the 1c site and In(2): the 4i site]. In(1) has tetragonal local symmetry, while the In(2) site is orthorhombic. To perform a comprehensive examination of the two-fluid model, K and T_1 have been measured at both In sites with applied magnetic field $H \parallel c$ axis([001]) and $H \parallel a$ axis([100]). The NMR shifts K, nuclear quadrupole frequencies ν_Q and asymmetry parameters η have been determined using an exact diagonalization method for the case of $I=9/2$. T_1 is determined for the central transition ($m=1/2 \leftrightarrow -1/2$) recovery which is well fitted with calculated curves for $I=9/2$, $\eta=0$ for In(1) and $\eta=0.461$ for In(2) sites. Since $\omega_n \gg \nu_Q$, the anisotropy of T_1 can be determined correctly even for $\eta \neq 0$ case. The linewidth of the In NMR spectrum is small ~ 0.1 kOe (inset to Fig. 1), guaranteeing a precise determination of K and T_1.

Since the principal axis $\vec{n}_{ZZ}$ of the electric field gradient tensor is perpendicular to the (100) plane at the In(2) site, two different In(2) site orientations (*i.e.* for K and T_1) are observed when $H \parallel a$: the In(2a) site ($H \perp \vec{n}_{ZZ}$) and the In(2b) site ($H \parallel \vec{n}_{ZZ}$). For the other cases, K and T_1 are determined uniquely. It should be noted that our results for ν_Q and η at both In(1) and In(2) sites are precisely consistent with previous zero-field nuclear quadrupole resonance (NQR) results.[7,8]

Figure 1 shows the T dependence of the static susceptibility χ_α ($\alpha=a,c$ for $H \parallel a,c$, respectively). The observed rapid increase in χ_c below 15 K is confirmed as an intrinsic property since it is independent of applied magnetic field. The inset to Fig. 2(a) shows the T dependence of $K_{i,\alpha}$, where $i=1$ and 2 for the In(1) and In(2) sites; $\alpha=a$ and c for $H \parallel a$ and $H \parallel c$ at the In(1) site; $\alpha=a$, b and c for $H \perp \vec{n}_{ZZ}$ [In(2a)], $H \parallel \vec{n}_{ZZ}$ [In(2b)], and $H \parallel c$ at the In(2) site, respectively.

In order to compare Knight shifts with the static susceptibility, plots of $K_{i,\alpha}$ versus χ_α (K-χ plots) are presented in Fig. 2. Above ~ 60 K, nearly linear relations between $K_{i,\alpha}$ and χ_α (here $\chi_a=\chi_b$) are found for all cases. From the nearly linear high-temperature slopes, the transferred hyperfine cou-

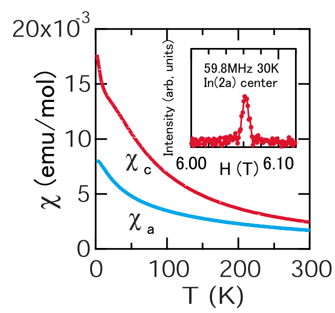

FIG. 1. (Color online) T dependence of the static susceptibility χ_α ($\alpha=a$-axis and $\alpha=c$-axis). The inset shows the In(2a) NMR spectrum for the central ($m=1/2 \leftrightarrow -1/2$) transition. All other spectra are similar to this.

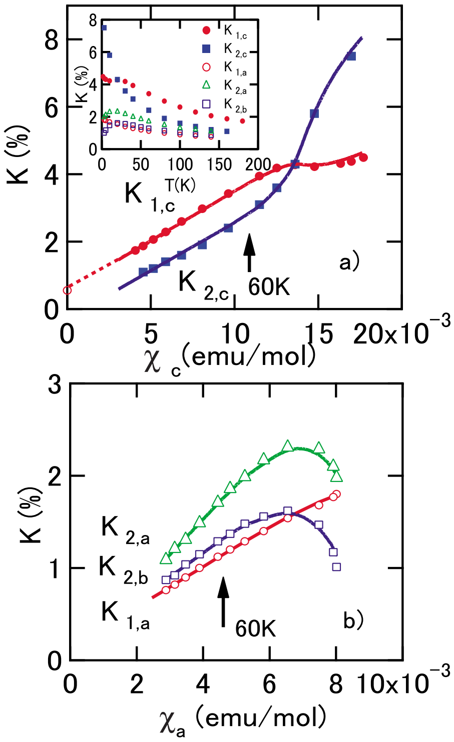

FIG. 2. (Color online) Knight shift $K_{i,\alpha}$ versus static susceptibility χ_α plots for the In(1) ($i=1$) and In(2) ($i=2$) sites, a) for $H \parallel c$-axis ($\alpha=c$), and b) for $H \parallel a$-axis ($\alpha=a,b$). The non-linear behavior looks different for $H \parallel c$-axis and $H \parallel a$-axis, since χ_c increases with decreasing T more rapidly than χ_a (Fig. 1). $K_{1,c}^{ori}$ is indicated as an example of $K_{i,\alpha}^{ori}$. Inset: T-dependence of $K_{i,\alpha}$. Solid lines drawn are calculated curves based on Eq. (1). Size of symbols represents experimental error.

pling constants $A_{i,\alpha}^{ht}$ can be estimated (Table I).

With decreasing T, the K-χ plots deviate markedly from linearity. It is interesting to note that all such deviations scale reasonably well with the function χ_α^{hf}, as in

$$K_{i,\alpha}^{spin} \equiv K_{i,\alpha} - K_{i,\alpha}^0 = A_{i,\alpha}^{ht}\chi_\alpha + C_{i,\alpha}\chi_\alpha^{hf}, \qquad (1)$$

where $K_{i,\alpha}^{spin}$ is the T-dependent spin Knight shift, the $C_{i,\alpha}$ are certain constants, and $K_{i,\alpha}^0$ is a small constant Knight shift which may be orbital in origin (see below). Furthermore, the T dependence of χ_α^{hf} is very nearly proportional to the density of states $D(T)$ given by the results of dynamical mean-field-theory (DMFT) calculations,[9] $\chi_\alpha^{hf} \propto D(T)$. We have chosen the $C_{i,\alpha}$ so that χ_α^{hf} fits with the enhanced Pauli paramagnetic susceptibility estimated from $D(T)$, thus

TABLE I. Transferred hyperfine coupling constants $A_{i,\alpha}^{ht}$ and $C_{i,\alpha}$ (in kOe/μ_B) for $K_{i,\alpha}$.

	$\alpha=a$	$\alpha=b$	$\alpha=c$
$A_{1,\alpha}^{ht}$	12.2 ± 0.3		16.4 ± 0.3
$A_{2,\alpha}^{ht}$	21.5 ± 0.3	14.5 ± 0.3	15.9 ± 0.3
$C_{1,\alpha}$	-0.65 ± 0.03		-3.1 ± 0.1
$C_{2,\alpha}$	-6.5 ± 0.2	-6.5 ± 0.2	9.3 ± 0.3

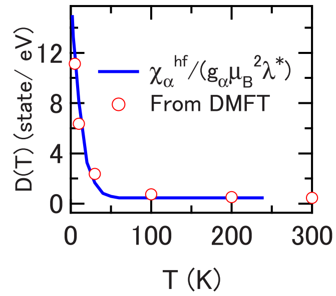

FIG. 3. (Color online) Open circles: Density of states $D(T)$ from DMFT calculation (Ref. 9). Solid line is T-dependence of $\chi_\alpha^{hf}/g_\alpha\mu_B^2\lambda^*$ which is compared with all K-χ plots using Eq. (1).

$\chi_\alpha^{hf} \simeq 0.5 g_\alpha^2 \mu_B^2 \lambda^* D(T)$ ($g_a=g_b=1.31$, $g_c=1.92$) as shown in Fig. 3, where g_α is determined by the condition that $0.5 g_\alpha^2 \mu_B^2 \lambda^* D(T)$ agrees with χ_α at $T \sim 0$ K, the mass enhancement factor λ^* is ~ 10.[10] The corresponding values of $C_{i,\alpha}$ (hyperfine coupling dimensions) so determined are given in Table I. The anisotropy of the $A_{i,\alpha}^{ht}$ and $C_{i,\alpha}$ parameters is due to the anisotropy of the transferred hyperfine couplings. The small constant $K_{i,\alpha}^0$ is determined by $K_{i,\alpha}^0 = K_{i,\alpha}^{ori} - C_{i,\alpha} g_\alpha \mu_B^2 \lambda^* D^{ht} \simeq K_{i,\alpha}^{ori}$, where $K_{i,\alpha}^{ori}$ is the extrapolated value of $K_{i,\alpha}$ at $\chi_\alpha=0$ [e.g., see Fig. 2(a)], and $D^{ht} \simeq 0.4$ (states/eV) is the high-temperature limiting value of $D(T)$.[9] In Fig. 2 calculated (solid) lines based on Eq. (1) are presented, showing good agreement with measured shift curves.

These findings are essentially the same as observations on many heavy-fermion compounds based on the two-fluid model.[3,4] In the frame of the two-fluid model,[3] the two components of NMR shift can be expressed as $K_{i,\alpha}^{spin} = A_{i,\alpha}^{ht}[1-f(T)]\chi_\alpha^{SL} + (A_{i,\alpha}^{ht} + C_{i,\alpha}) f(T)\chi_\alpha^{KL}$, where $[1-f(T)]\chi_\alpha^{SL}$ is the localized component susceptibility, $f(T)\chi_\alpha^{KL}$ is the heavy-fermion component susceptibility, and $\chi_\alpha \simeq [1-f(T)]\chi_\alpha^{SL} + f(T)\chi_\alpha^{KL}$. In fact the scaling function of the two-fluid model $f(T) \sim (1-T/T^*)^{1.5}$ ($T^* \sim 31$ K) is confirmed to be consistent with $D(T) \propto f(T)\chi_\alpha^{KL} \simeq \chi_\alpha^{hf}$ for $CeIrIn_5$.[3]

The present results show that all deviations scale to the unique function $D(T)$ for both In sites and all applied field orientations. Thus T^* is an isotropic "thermodynamical" quantity characterizing low-lying states of the system. The observed T^* is isotropic under magnetic field ~ 7 T although it was pointed out that an applied magnetic field may cause T^* to become anisotropic.[11] Indeed, recent NMR measurements suggest an isotropic nature for T^* in $CeCoIn_5$, except for the In(2) site.[12]

In addition to the two-fluid picture, the observed nonlinear K-χ relation can also be interpreted in a different way. If the heavy-fermion and localized components were merged into a single component, the nonlinear behavior could simply be seen as a modification of the hyperfine coupling for the total χ_α. In this "*merged, one-component*" case, the hyperfine coupling constant becomes T dependent

$$K_{i,\alpha}^{spin} = \{A_{i,\alpha}^{ht} + C_{i,\alpha}\chi_\alpha^{hf}/\chi_\alpha\}\chi_\alpha \equiv A_{i,\alpha}(T)\chi_\alpha, \qquad (2)$$

where the T-dependent hyperfine coupling constant $A_{i,\alpha}(T)$ asymptotically approaches $A_{i,\alpha}^{ht}$ at high T. In this description, the change $\Delta A_{i,\alpha}(T) \equiv C_{i,\alpha}\chi_\alpha^{hf}/\chi_\alpha$ is scaled to a proportion of the heavy-fermion component $\chi_\alpha^{hf}/\chi_\alpha \simeq g_\alpha \mu_B^2 \lambda^* D(T)/\chi_\alpha$. At the same time, the measured χ_α increases owing to the contribution from χ_α^{hf}. The scaling between $\Delta A_{i,\alpha}(T)$ and $D(T)$ indicates that the hybridization at the In site is modified due to heavy-fermion formation.

Concerning the relation between $A_{i,\alpha}(T)$ and $D(T)$, it is difficult to distinguish between Eqs. (1) and (2) on the basis of the K-χ result. In contrast, the T_1 results are quite useful for distinguishing them as described below.

We define subcomponents of the spin-lattice relaxation rates from fluctuations along the α axes: $R_{i,\alpha} \equiv \gamma_n^2 \Sigma_q A_{i,\alpha}^2 \chi''_\alpha(q,\omega_n)/\omega_n$, where $\chi''_\alpha(q,\omega_n)$ is the dynamical susceptibility tensor at frequency ω_n. $\chi''_\alpha(q,\omega_n)$ originates with the $4f$ moments on the tetragonal Ce sites and has components $\chi''_a(q,\omega_n) = \chi''_b(q,\omega_n)$ and $\chi''_c(q,\omega_n)$. $A_{i,\alpha}(q)$ is the transferred hyperfine coupling constant at the In sites. Effects of the q dependences of $A_{i,\alpha}(q)$ and $\chi''_\alpha(q,\omega_n)$ will be touched upon below. At the In(1) site, $1/(T_1T)_{H\|a} = R_{1,a}+R_{1,c}$, and $1/(T_1T)_{H\|c} = 2R_{1,a}$. At the In(2) site, $1/(T_1T)_{H\perp \vec{n}_{ZZ}} = R_{2,b}+R_{2,c}$; $1/(T_1T)_{H\|\vec{n}_{ZZ}} = R_{2,c}+R_{2,a}$; and $1/(T_1T)_{H\|c} = R_{2,a}+R_{2,b}$.[13]

Figures 4(a) and 4(b) show the T dependences of the $R_{i,\alpha}$ determined using the foregoing equations. As expected, the rate components increase with decreasing T along with χ_α. However, it is striking that $R_{1,c}$ and $R_{2,c}$ show contrasting T dependences at low T. Since the In sites probe transferred hyperfine fields from the Ce site, $R_{1,c}$ and $R_{2,c}$ should be proportional to each other if they correspond to unique magnetic fluctuations at the Ce site along the c axis. The observed discrepancy indicates that $R_{1,c}$ and $R_{2,c}$ probe the transferred hyperfine fields from the Ce moments differently.

We may try to explain this complex behavior on the basis of a two-fluid picture assuming no coherence between the two components. For such a case, $R_{i,\alpha}$ may be expressed as

$$R_{i,\alpha} \simeq R_{i,\alpha}^{incoh} \equiv (A_{i,\alpha}^{ht})^2[1-f(T)]\mathrm{Im}\,\chi_\alpha^{SL} + (A_{i,\alpha}^{ht} + C_{i,\alpha})^2 f(T)\mathrm{Im}\,\chi_\alpha^{KL} \quad (3)$$

with the normalized dynamical susceptibility $\mathrm{Im}\,\chi_\alpha^{SL,KL} \equiv \gamma_n^2 \chi_\alpha''^{SL,KL}(q,\omega_n)/\omega_n$. Based on Eq. (3) and Table I, the T dependence of $\mathrm{Im}\,\chi_\alpha^{SL}$ and $\mathrm{Im}\,\chi_\alpha^{KL}$ should be obtainable from $R_{1,\alpha}$ and $R_{2,\alpha}$ below $T^* = 31$ K without any adjustable parameters. However, as shown in Fig. 4(c), estimated values for $\mathrm{Im}\,\chi_c^{SL}$ and $\mathrm{Im}\,\chi_a^{KL}$ become negative, showing a peculiar T dependence. Furthermore, $\mathrm{Im}\,\chi_a^{KL}$ is not proportional to $\mathrm{Im}\,\chi_c^{KL}$ in spite of the fact that $\chi_a^{hf} \propto \chi_c^{hf}$. Thus, the "noncoherent" two-fluid picture does not appear to be physically viable.[14] However, we note that it has been reported that the NQR T_1 at the In(1) site in $\mathrm{CeCoIn_5}$ can be explained qualitatively using a noncoherent, two-fluid model if a particular T dependence of $\mathrm{Im}\,\chi_\alpha^{SL}$ is assumed.[12]

On the other hand for the merged one-component case, the normalized one-component dynamical susceptibility $\mathrm{Im}\,\chi_{i,\alpha} \equiv \gamma_n^2 \chi''_{i,\alpha}(q,\omega_n)/\omega_n$ can be simply estimated with

$$R_{i,\alpha} \simeq R_{i,\alpha}^{coh} \equiv A_{i,\alpha}(T)^2 \mathrm{Im}\,\chi_{i,\alpha}. \quad (4)$$

Figure 4(d) shows the T dependence of the four dissipative terms $\mathrm{Im}\,\chi_{i,\alpha}$ ($i=1,2$ $\alpha=a,c$) estimated in this fashion. In this limit, the $\mathrm{Im}\,\chi_{i,\alpha}$ all increase naturally with decreasing T. At high T, relations naturally expected for transferred hyperfine fields from a unique Ce site, i.e. $\mathrm{Im}\,\chi_{1,c} \sim \mathrm{Im}\,\chi_{2,c}$, and $\mathrm{Im}\,\chi_{1,a} \sim \mathrm{Im}\,\chi_{2,a}$, are confirmed.[15] The consistent results we find with Eq. (4) indicate that it is not necessary to introduce a two-component dynamical susceptibility. Although it is not clear how localized and heavy-fermion components merge in the one-component picture, the present results suggest that they merge smoothly to form a single dynamical entity, while treating them as independent entities yields apparently unphysical results.

Above 100 K, the product $T \times \mathrm{Im}\,\chi_{i,\alpha}$ becomes asymptotically constant, indicating that the excitation behavior approaches that of localized $4f$ moment fluctuations. Below 30 K, in contrast to high T, $\mathrm{Im}\,\chi_{2,a}$ is larger than $\mathrm{Im}\,\chi_{1,a}$. This can be explained if fluctuations develop at the antiferromagnetic wave vector Q: $\chi''_\alpha(Q,\omega_n) > \chi''_\alpha(0,\omega_n)$ at low T. Such fluctuations would cancel completely at the In(1) site but not at the In(2) site because of different q dependences for

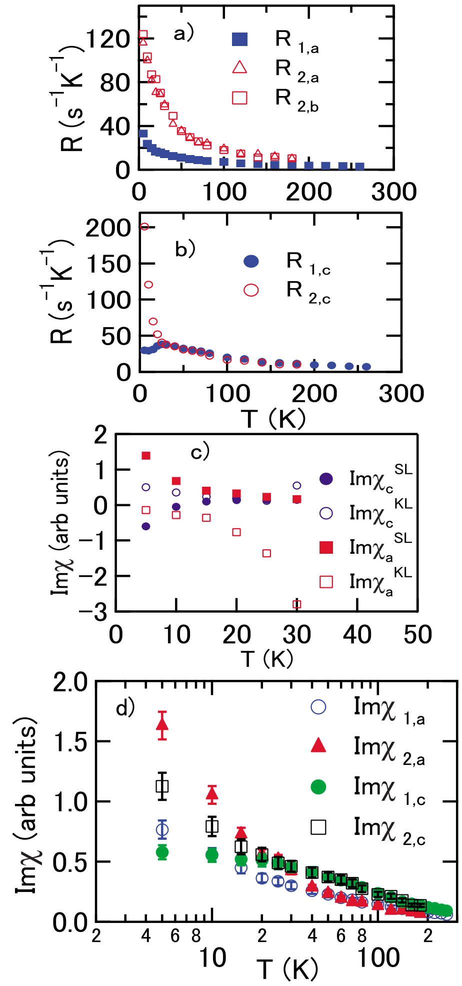

FIG. 4. (Color online) (a) T-dependence of spin-lattice relaxation rates along a-axis: $R_{1,a}$ $R_{2,a}$ and b-axis: $R_{2,b}$. b) T-dependence of spin-lattice relaxation rates along c-axis: $R_{1,c}$ and $R_{2,c}$. c) T-dependence of $\mathrm{Im}\,\chi_\alpha^{SL}$ and $\mathrm{Im}\,\chi_\alpha^{KL}$ on assumption of an incoherent two-fluid description estimated using Eq. (3). Size of symbols represents an experimental error. d) T-dependence of $\mathrm{Im}\,\chi_{i,\alpha}$ using Eq. (4) based on a merged one-component description.

$A_{i,\alpha}(q)$.[16] Meanwhile, anisotropy of the fluctuations $\text{Im}\,\chi_{i,a}/\text{Im}\,\chi_{i,c}$ also develops, indicating that $CeIrIn_5$ is approaching an antiferromagnetically ordered state with an ordered moment in the (001) plane. Such an ordered state was in fact observed for $CeRhIn_5$.[17] The same anisotropy is also enhanced at low T in $CeCoIn_5$.[18]

The relation $\text{Im}\,\chi_{1,a} \propto (T+8\ \text{K})^{-3/4}$ is obtained below 100 K as well, consistent with a previous NQR T_1 result for the In(1) site.[7,8] However, the exponent $-3/4$ is not universal since it has a value ~ -1 for $\text{Im}\,\chi_{2,a}$ while it is difficult to define such an exponent for $\text{Im}\,\chi_{i,c}$. The anisotropy of the fluctuations is considered to characterize the nature of the magnetism in any particular case.

The estimated anisotropy of the dynamical susceptibility ($\text{Im}\,\chi_a > \text{Im}\,\chi_c$) is opposite to that of the static susceptibility χ_α ($\chi_c > \chi_a$) at low T, which may be due to q-dependent exchange interactions. Previously we have pointed out that this XY-type of magnetic fluctuation anisotropy, i.e. $\text{Im}\,\chi_a \gg \text{Im}\,\chi_c$, is favorable for antiferromagnetic d-wave superconductivity.[19] The present example reinforces this hypothesis.

In conclusion, the nonlinear K-χ relation is well described phenomenologically by the two-fluid model in $CeIrIn_5$. However, this could also be interpreted in a one-component picture, where the transferred hyperfine coupling is modified by a contribution scaled to the quasiparticle density of states. The latter effect is seen clearly in the shift results. Such a description suggests that a one-component dynamical susceptibility can successfully describe the magnetic fluctuations in a heavy-fermion system. How such a one-component dynamical susceptibility can be reconciled with a static susceptibility apparently composed of two contributions remains to be resolved.

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[15] For $\text{Im}\,\chi_{2,b}$, a relation $\text{Im}\,\chi_{2,b} \simeq (A_{2,a}^{ht}/A_{2,b}^{ht})^2 \text{Im}\,\chi_{2,a}$ is obtained, indicating that the generally expected relation $\text{Im}\,\chi_{2,b} \simeq \text{Im}\,\chi_{2,a}$ may be modified due to off-diagonal contributions to $A_{2,b}^{ht}$. Actually, the effective $A_{2,b}^{ht}$ for spin-lattice relaxation is considered to be similar to $A_{2,a}^{ht}$.

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