Origin for the enhanced copper spin echo decay rate in the pseudogap regime of the multilayer high-$T_c$ cuprates

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We report measurements of the spin echo decay for the inner layer Cu site of the triple layer cuprate, $\text{Hg}_2\text{Ba}_2\text{Ca}_2\text{Cu}_{3}\text{O}_8$ ($T_c=126$ K) in the pseudogap $T$ regime below $T_{pg} \sim 170$ K and the corresponding analysis for their interpretation. As the field alignment is varied, the shape of the decay change from Gaussian ($H_0 \parallel c$) to single exponential ($H_0 \perp c$). The latter characterizes the decay caused by the fluctuations of adjacent Cu nuclear spins caused by their interactions with electron spins. The angular dependence of the second moment ($T^{2M}_2 \equiv \langle \Delta \omega^2 \rangle$) deduced from the decay curves indicates $T^{2M}_2 \parallel H_0 \parallel c$, which is identical to $T^{2G}_2$ ($T^{2G}_2$ is the Gaussian component), is substantially enhanced, as seen in the pseudogap regime of the bilayer systems. Comparison of $T^{2M}_2$ between $H_0 \parallel c$ and $H_0 \perp c$ indicates that this enhancement is caused by electron spin correlations between the inner and the outer CuO$_2$ layers. These results provide the answer to the long-standing controversy regarding the opposite $T$ dependences of $(T_1T)^{-1}$ and $T^{2G}_2$ in the pseudogap regime of bi- and trilayer systems.

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data by exp(−2τ/T_{1R}) \textsuperscript{14} with T_{1R} obtained as follows [17]. The general form of T_{1R}^{-1} is given by

\[(T_{1R})_z^{-1} = \{I(I + 1) - 1/4\}(W_x + W_y) + W_z, \quad (1)\]

where W_{\gamma} (\gamma=x, y, z) is from the spin fluctuations in the γ direction and z is the quantization axis (\| H_0 ). In the same notation, (T_1)_z^{-1} is given by (T_1)_z^{-1} = W_x + W_y. Hence, for an arbitrary θ,

\[\left[T_{1R}(\theta)\right]^{-1} = \{I(I + 1) - 1/4\}[T_1(\theta)]^{-1}\]

\[+\left[T_1(90^\circ - \theta)\right]^{-1} - 0.5\left[T_1(0^\circ)\right]^{-1}, \quad (2)\]

where the relation W_{\gamma} = W_{\gamma} is used (The subscripts a, b correspond to the crystalline axes). From the anisotropy of (T_1(T))^{-1} in Fig. 2(b), \left[T_{1R}(\theta)\right]^{-1} is calculated.

Figure 2(a) shows that the shape of the decay curve changes from Gaussian at 0° to single exponential (Lorentzian spectrum) at 90°. These data are fitted by the function,

\[M(2\tau) = M_0 \exp \left[ -\frac{2\tau}{T_{2L}} - \frac{1}{2} \left( \frac{2\tau}{T_{2G}} \right)^2 \right] \quad (3)\]

with M_0, T_{2L}, and T_{2G} as free parameters. The angular dependences of T_{2L}^{-1} and T_{2G}^{-1} thus obtained are shown in Fig. 2(c). For 0° ~ 20°, T_{2L}^{-1} is nearly zero while T_{2G}^{-1} is zero at 80° and 90°. In between, the decay curve crosses over between these two extremes. This change is a result of the NMR line narrowing caused by the following mechanism. At finite T the effective interaction between adjacent nuclear spins is reduced because of rapid spin flips driven by the hyperfine interaction with the electrons, which averages out the nuclear fields at adjacent nuclear sites. As a result, the central part of the NMR line is narrowed and the spectrum approaches a Lorentzian rather than a Gaussian shape [17, 18]. The importance of the effect depends on the ratio between the two time scales T_1 and T_{2M} = (\Delta \omega^2)^{-1/2}, where (\Delta \omega^2) is the homogeneous second moment of the NMR absorption. The former and the latter characterize the time scales of the nuclear spin fluctuations and the echo decays, respectively.

There are two limiting cases where analytical forms for the decays are known. One is the static limit (T_1/T_{2M} \gg 1), where nuclear spins do not change their states between pulses or a pulse and an echo because of the relatively long T_1. Consequently, the contributions from unlike-spins are canceled out at the time of the echo and only like-spins contribute to the echo decay. The decay in this case is described by a Gaussian [2].

\[M(2\tau) = M_0 \exp \left[ -\frac{1}{2} \left( \frac{2\tau}{T_{2M}} \right)^2 \right] f(2\tau), \quad (4)\]

where f(2\tau) is the correction for the narrowing effect [13] and in the static limit, f(2\tau) = 1. The rate T_{2M}^{-1} in this case is usually referred to as the Gaussian rate, T_{2G}^{-1}.

The second is the narrowed limit (T_1/T_{2M} \leq 1), where nuclei are fluctuating during the echo sequence. The decay is characterized by a single exponential (Lorentzian spectrum) [17, 18],

\[M(2\tau) = M_0 \exp \left[ -2\tau \left( \frac{63T_1}{63T_{2M}^2} + \frac{65T_1}{65T_{2M}^2} \right) \right] \quad (5)\]

where \alpha T_{2M}^{-2} = \alpha (\Delta \omega^2) is the contribution from the α-nuclei (α=63, 65) to the second moment of 63Cu, and \alpha T_1 is the spin-lattice relaxation time in the α-site. Note that 63T_{2M} corresponds to T_{2G} in the static limit. Here, 65Cu also contributes to the 65Cu decay because they lose their memories of the initial states during the echo sequence due to the fluctuation effect, so that their contributions are not canceled out at the time of the 63Cu echo.

In the present case, the transition from the static to the narrowing regime is caused by the large anisotropies of T_1^{-1} and T_{2M}^{-1}. As will be shown later, T_{2M}^{-1} decreases

FIG. 1: Absorption spectra for the central transition of 63Cu at 135 K as a function of θ. The triangles show the part of the spectra used for the T_2 measurements. Inset: Schematic view of the CuO$_4$ layers.

FIG. 2: Angular dependences of (a) the echo decay curve, (b) \((T_1T)^{-1}\), (c) \(T_{2G}^{-1}\) (●) and \(T_{2L}^{-1}\) (Δ) in Eq. 3 and (d) \(T_{2M}^{-1}\) deduced from \(T_{2G}^{-1}\) (●) and \(T_{2L}^{-1}\) (Δ) at 135 K. The dashed curve in (d) is given by Eq. 5.
by 3.3 from $\theta = 0^\circ$ to $90^\circ$, while $T_{1}^{-1}$ increases by 2.1. Also, there is a qualitative correspondence between the transition of the decay curve in Fig. 2(a) and the simulations by Walstedt et al. for the various values of $T_{1}/T_{2M}$ (Fig. 4 of Ref. [13]). The angular dependence of $T_{2M}^{-2}$ deduced from either $T_{2G}$ or $T_{2L}$ in Fig. 2(c) is shown in Fig. 2(d), where $T_{2M} = T_{2G}$ while Eq. (3) is used to obtain $^{63}T_{2M}$ from $T_{2L}$ along with the relations,

$$^{63}T_{1} = (65\gamma)^{2} \cdot 65^{\frac{1}{2}} T_{1}, \quad (6)$$

$$^{65}T_{1} = (65\gamma)^{2} \cdot 65^{\frac{1}{2}} T_{1}, \quad (7)$$

where $^{63}T_{1}$ is the form factor when $T_{2G}$ or $T_{2L}$ is presented by $\chi_{ij}(q)$, which is the natural abundance for the isotope $^{63}T_{1}$.

The angular dependence of $T_{2M}^{-2}$ in Fig. 2(d) is consistent with that of the hyperfine coupling constant. In the detuned limit, the flip-flop term $(F_{11}^{T}F_{11}^{T})$ in the nuclear Hamiltonian is ineffective because of the mismatch in the Zeeman energies between adjacent nuclei. Hence, $T_{2M}$ is given only by the z-component term $(F_{z}^{T}F_{z}^{T})$, so that $[T_{2M}(\theta)]^{-2} \propto (\chi(Q))^{2} \cdot \sum_{q} F_{q}(\theta)^{2}$, where $F_{q}(\theta)$ is the form factor when $H_{0}$ is in the $\theta$ direction [3]. Here, we assume that the $q$-dependence of $\chi(Q)$ around $Q = (\pi, \pi)$ is weaker than that of $F_{q}$, so that $\chi(Q)$ is represented by $\chi(Q)$ and taken out of the $q$-summation. We further assume that $\sum_{q} F_{q}(\theta)^{2}$ is proportional to $(F_{q}(\theta))^{2}$ at each $\theta$. Since the $\theta$ dependence of $F_{q}(\theta)$ is $(A(3\cos^{2}\theta - 1) + B)^{2}$ where $A$ and $B$ are constants, the anisotropy of $T_{2M}^{-2}$ is given by

$$[T_{2M}(\theta)]^{-2}/[T_{2M}(90^\circ)]^{-2} = (\sin^{2}\theta + \xi \cos^{2}\theta)^{4}, \quad (8)$$

where

$$\xi \equiv \left[ \frac{\sum_{q} F_{q}(0^\circ)^{2}}{\sum_{q} F_{q}(90^\circ)^{2}} \right]^{1/4} \approx \left[ \frac{F_{q}(0^\circ)}{F_{q}(90^\circ)} \right]^{1/2}. \quad (9)$$

The value of $\xi$ can be estimated from the anisotropy of $T_{1}^{-1}$. Since $\chi(Q) \gg \chi(0)$ in this system, $(T_{1})^{-1} \propto \{F_{x}(Q) + F_{y}(Q)\}[4]$. Hence, $\xi$ is given by,

$$\xi \approx \left[ \frac{F_{q}(0^\circ)}{F_{q}(90^\circ)} \right]^{1/2} \approx \left[ \frac{(T_{1}(90^\circ))^{-1}}{(T_{1}(0^\circ))^{-1} - 1} \right]^{1/2}. \quad (10)$$

From Fig. 2(b), $(T_{1}(90^\circ))^{-1}/(T_{1}(0^\circ))^{-1} = 2.1$, so that $\xi = 1.79$. The dashed curve in Fig. 2(d) shows the anisotropy of $T_{2M}$ obtained from Eq. (3) with $(T_{2M}(90^\circ))^{-2}$ as a single adjustable parameter. It has the same tendency as the angular dependence of $T_{2G}$ in spite of some assumptions.

The analysis at $0^\circ$, $10^\circ$ and $70^\circ$ is not straightforward because of the overlap with the Cu(2) line. Since $T_{2M}$ at Cu(2) is expected to be smaller than that of Cu(1) by a factor of $0.75$ [4], the overlapped Cu(2) line reduces $T_{2M}^{-2}$, however, is not reduced at $10^\circ$, and is significantly enhanced at $0^\circ$. At $70^\circ$, the shape of the decay curve itself is quite different from those at $60^\circ$ and $80^\circ$. Below, we show that these features can be attributed to the interlayer spin correlations, which have the effect of enhancing $T_{2M}$ [4, 8].

![FIG. 3: Schematic view of the intra (i = j) and interlayer (i ≠ j) spin susceptibilities (χij(q)) in the trilayer system.](image)

Consider the echo decay process at $\theta = 0^\circ$. As seen in Fig. 1, the Cu(1) and Cu(2) lines are situated close to each other, so that not only Cu(1) but also a part of Cu(2) nuclei are excited, which also act as like-spins for the Cu(1) nuclei in the echo decay process. On the other hand, the intensity of the echo is obtained by integrating only the Cu(1) part of the FT spectrum of the echo (shaded part of the spectrum in Fig. 1), so that only the Cu(1) nuclei contribute to the intensity of the decay curve in Fig. 2(a). This is the same situation as that in the SEDOR experiment where the $\pi$-pulses for like- and unlike-spins are applied simultaneously. Since all the like-spins contribute to the Gaussian decay in the static limit, $[T_{2M}(0^\circ)]^{-2}$ is given by

$$[T_{2M}(0^\circ)]^{-2} \propto \sum_{q} [F_{q}(0^\circ)\{\chi(11)(q) + 4\epsilon\chi(12)(q)\}], \quad (11)$$

where, $\chi(11)$ and $\chi(12)$ are the intra- and interlayer spin susceptibilities associated with the auto- and cross-correlations within or between layers indicated in Fig. 2. The second term in Eq. (11) corresponds to the contribution due to the interlayer correlations. The ratio of the excited Cu(2) nuclei ($\epsilon$) is estimated to be about 0.6.

At the angles where the two Cu lines are separated from each other, the second term in Eq. (11) does not appear in the echo decay process, whereas at $10^\circ$ and $70^\circ$, both the Cu(1) and Cu(2) nuclei are excited and observed, so that the second term in Eq. (11) also appears, which enhances $T_{2M}^{-2}$. This enhancement increases $T_{1}/T_{2M}$, and brings the situation at $70^\circ$ closer to the static limit, resulting in the appearance of the Gaussian component in the decay curve. At $10^\circ$, a cancelation may occur between the reduction due to the overlapped Cu(2) line and the enhancement due to the interlayer spin correlations.

Figure 2(b) shows the $T$ dependences of $T_{2M}^{-2}$ at $0^\circ$ and $90^\circ$, which are quite different from each other; i.e., while $[T_{2M}(90^\circ)]^{-2}$ starts to decrease at $T_{pg}$ as does $(T_{1}T)^{-1}$ shown in Fig. 2(a), $[T_{2M}(0^\circ)]^{-2}$ continues to grow down to $T_{c}$. This difference is caused by the $\chi(12)$ term in Eq. (11). Provided again that the $q$-dependences of $\chi(12)(q)$ around $Q$ are weaker than that of $F_{q}$, $T_{2M}^{-2}$ at $0^\circ$ and $90^\circ$
can be rewritten as \[ (13), \]

\[
\frac{[T_{2M}(0^\circ)]^{-2}}{[T_{2M}(90^\circ)]^{-2}} \propto \left\{ \chi^{11}(Q) + 4\epsilon \chi^{12}(Q) \right\}^2 \cdot \sum_q \{F_q(0^\circ)\}^2,
\]

\[
\frac{[T_{2M}(90^\circ)]^{-2}}{[T_{2M}(0^\circ)]^{-2}} \propto \chi^{11}(Q)^2 \cdot \sum_q \{F_q(90^\circ)\}^2.
\]

(12)

Here, we define the ratio \( \rho \) by,

\[
\rho = \frac{[T_{2M}(0^\circ)]^{-2}}{[T_{2M}(90^\circ)]^{-2}} \cdot \xi^{-4} = \left[ 1 + \frac{4\epsilon \chi^{12}(Q)}{\chi^{11}(Q)} \right]^2 ,
\]

(13)

which gives,

\[
\chi^{12}(Q)/\chi^{11}(Q) = (\sqrt{\rho} - 1)/4\epsilon .
\]

(14)

The \( T \) dependence of \( \chi^{12}(Q)/\chi^{11}(Q) \) calculated from \( \rho \) is shown in Fig. \( (14) \), where \( \xi \) is adjusted so that \( \rho = 1 \) at high \( T \). One can see that \( \chi^{12}(Q)/\chi^{11}(Q) \) rapidly increases in the pseudogap \( T \) region, indicating that \( \chi^{12}(Q) \) rapidly grows there. This is consistent with the SEDOR results \( (15) \) and the theoretical calculations \( (16) \) (17). Note that \( \chi^{12}(Q)/\chi^{11}(Q) \equiv T_{2S}/T_{2G} \) where \( T_{2S} \) is the SEDOR decay time between the sites on the different layers. From Fig. \( (18) \), we find that,

\[
\chi^{12}(Q)/\chi^{11}(Q) = 0.28 ,
\]

(15)

at \( T_c \), while \( T_{2S}/T_{2G} \) was reported to be about 0.25 at \( T_c \) in \( Y_2Ba_4Cu_7O_{15-\delta} \). The consistent explanation for both \( T_{2S}/T_{2G} \) and \( T_{2S} \) indicates that they can be interpreted on the same basis of the interlayer spin correlations.

In conclusion, we have investigated the anisotropy of the spin echo decay in the inner Cu site of the trilayer cuprate \( \text{Hg}_0.8\text{Re}_{0.2}\text{Ba}_2\text{Cu}_3\text{O}_8 \) to obtain the anisotropy of the second moment \( \chi^{12}(Q) \). Comparison between the data at 0° and 90° shows the rapid growth of \( \chi^{12}(Q) \) in the pseudogap regime. Since this is a common effect in multilayer systems, we conclude that the opposite \( T \) dependences between \( (T_1T)^{-1} \) and \( T_{2G}^{-1} \) observed in the pseudogap regime of bilayer systems are caused by interlayer spin correlations. The UCLA part of the work was supported by NSF Grant DMR-0072524.

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