Coexistence of superconductivity and antiferromagnetism in multilayered high-T_c superconductor HgBa$_2$Ca$_4$Cu$_5$O$_y$: Cu-NMR study

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We report a coexistence of superconductivity and antiferromagnetism in five-layered compound HgBa$_2$Ca$_4$Cu$_5$O$_y$ (Hg-1245) with $T_c = 108$ K, which is composed of two types of CuO$_2$ planes in a unit cell; three inner planes (IP's) and two outer planes (OP's). The Cu-NMR study has revealed that the optimally doped OP undergoes a superconducting (SC) transition at $T_c = 108$ K, whereas the three underdoped IP's do an antiferromagnetic (AF) transition below $T_N \sim 60$ K with the Cu moments of $\sim (0.3$–$0.4) \mu_B$. Thus bulk superconductivity with a high value of $T_c = 108$ K and a static AF ordering at $T_N = 60$ K are realized in the alternating AF and SC layers. The AF-spin polarization at the IP is found to induce the Cu moments of $\sim 0.02 \mu_B$ at the SC OP, which is the AF proximity effect into the SC OP.

I. INTRODUCTION

There remain a number of underlying issues to be resolved in high-T_c cuprates. One of underlying issues is an interplay of antiferromagnetism and superconductivity in the antiferromagnetic (AF)–superconducting (SC) phase boundary, near vortex cores under magnetic field, and AF-SC alternate layered structures. Lake *et al.* reported that the AF correlations are induced in vortex cores and extend over the cores into the SC region in La$_{2-x}$Sr$_x$CuO$_4$ under the magnetic field, that is, an AF proximity effect into SC state.[1] This result is supported by some theoretical approaches.[2,3] In these theoretical predictions based on the SO(5) symmetry model,[4] each AF and SC fluctuation can extend into other region, when both the states come across. Recently, however, Bozovic *et al.* showed that the superconductivity does not mix into the AF insulator in the superconductor-insulator-superconductor heterostructures realized by stacking each layer of SC La$_{1.85}$Sr$_{0.15}$CuO$_4$ and AF La$_2$CuO$_4$.[5] Thus, these issues have not been settled yet.

Multilayered high-T_c cuprates, which have more than three CuO$_2$ planes in a unit cell, exhibit very unique magnetic and SC properties because they include two types of CuO$_2$ planes. As indicated in Fig. 1, an outer CuO$_2$ plane (OP) has a pyramidal five-oxygen coordination, whereas an inner plane (IP) has a square four-oxygen one. Note that the IP* is the middle plane of the three IP's as shown in the figure. Nuclear-magnetic-resonance (NMR) experiments revealed that the OP and the IP differ in the doping level.[7–10] We reported unusual magnetic and SC characteristics in multilayered CuO$_2$ planes in Hg and Cu-based high-T_c cuprates through ^{63}Cu-NMR measurements.[11] The Knight shift (^{63}K) at the OP and the IP exhibits different characteristic temperature (T) dependence, consistent with its own doping level. It was shown that the doping level N_h(OP) at the OP is larger than N_h(IP) at the IP for all the systems and its difference $\Delta N_h = N_h$(OP)$ - N_h$(IP) increases as either a total carrier content or n increases. At ΔN_h's exceeding a critical value, the respective SC transitions do not simultaneously set in at the IP and the OP.[12] Some theoretical approaches predict the effect induced by the carrier inhomogeneity.[13,14]

In this paper, we report ^{63,65}Cu-NMR study on Hg-1245 which evidences a coexistence of AF order at the IP's and IP* and bulk superconductivity at the OP. Note that the IP* is the middle plane of the three IP's as shown in Fig. 1. Measurements of the Knight shift ^{63}K, the nuclear-spin-lattice-relaxation rate ($1/T_1$), and the internal field (H_{int}) of ^{63,65}Cu have revealed that the OP undergoes a bulk SC transition below $T_c = 108$ K and the IP* and IP order antiferromagnetically below $T_N \sim 60$ K with the Cu moments of $0.37 \mu_B$ and $0.30 \mu_B$, respectively.

II. EXPERIMENTAL DATA AND DISCUSSION

Polycrystalline sample was prepared by the high-pressure synthesis technique as described elsewhere.[15] Powder x-ray-diffraction experiment indicates that the sample consists of almost a single phase, but includes a small fraction of Hg-1234.[15] A SC transition temperature of $T_c = 108$ K was determined from an onset T below which diamagnetic signal appears in dc susceptibility as shown in Fig. 2(a). For NMR measurements, the powdered sample was aligned along the c axis at $H = 16$ T. The NMR experiment was performed by the conventional spin-echo method at 174.2 MHz ($H \sim 15.3$ T).

Figure 2(b) shows NMR spectra for $H \perp c$. At 200 K, two well-separated peaks arise from the OP and the IP. The assignment of NMR spectrum to the OP and the IP was already reported in the previous literatures.[11,16] The NMR spectrum at the IP exhibits a sharper spectral width with a smaller
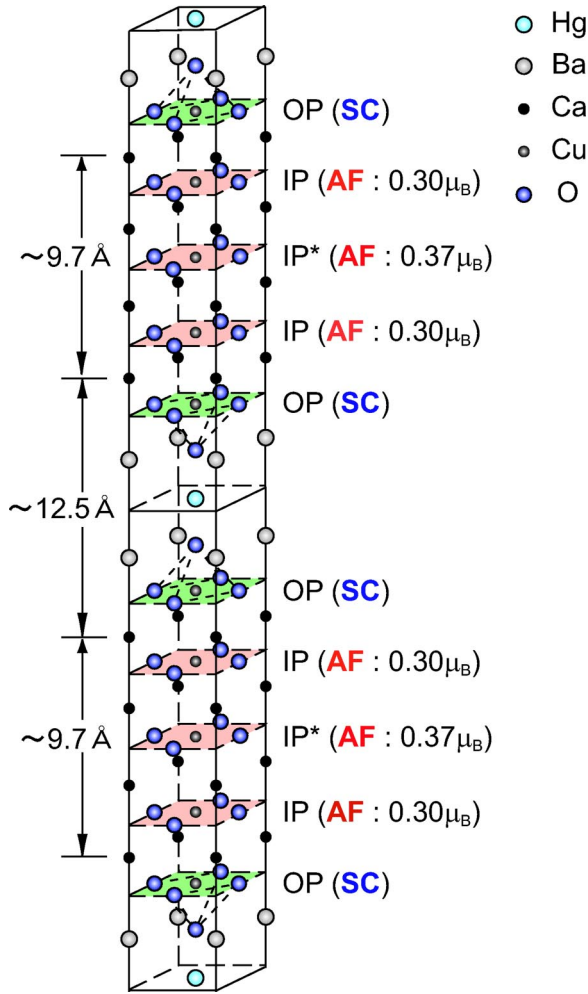
FIG. 1. (color on-line) The crystal structure of Hg-1245 ($a = 3.850$ Å, $c = 22.126$ Å) (Ref. 6). The OP undergoes the SC transition at $T_c = 108$ K, whereas the three underdoped IP's do an AF transition below $T_N \sim 60$ K with the respective Cu moments of $\sim 0.30 \mu_B$ and $0.37 \mu_B$ at the IP and the IP*.
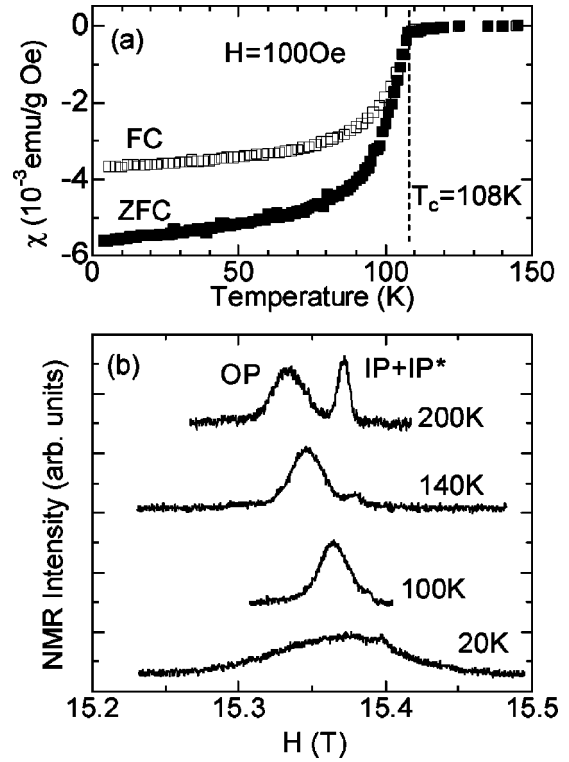

FIG. 2. (a) T dependence of dc susceptibility with field cooling (FC) and zero-field cooling (ZFC). A clear diamagnetic signal can be observed below $T_c = 108$ K. (b) The NMR spectra for $H \perp c$ at $T = 200$, 140, 100, and 20 K. The NMR signals at the IP and the IP* disappear below ~ 150 K. The OP's signal becomes significantly broader at temperatures lower than $T_N \sim 60$ K.
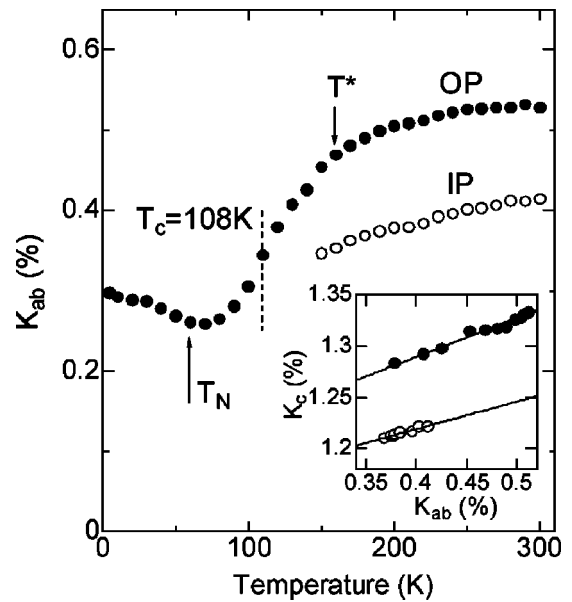

FIG. 3. T dependence of Knight shift K_{ab} for $H \perp c$. The inset of Fig. 3(b) shows K_{ab} vs K_c plots at the IP's and the OP with the temperature as an implicit parameter. These plots allow us to estimate the supertransferred hyperfine-coupling constants $B(\text{IP}) \sim 61$ kOe/μ_B and $B(\text{OP}) \sim 74$ kOe/μ_B, respectively.

Knight shift than those at the OP. The spectral width at the IP is estimated to be ~ 50 Oe for $H \| c$, comparable to the ~ 60 Oe for YBa$_2$Cu$_3$O$_7$ under $H \sim 15$ T, which is the narrowest among high-T_c cuprates to date. This ensures that the IP is rather homogeneously doped. The spectra at the IP and the IP* overlap each other, suggesting that their local doping levels are not so much different. The NMR signals at the IP and the IP* disappear due to their short relaxation time below $T \sim 150$ K.

Figure 3 indicates the T dependence of $^{63}K_{ab}$ at the IP's and the OP for $H \perp c$. In general, $K(T)$ consists of the T-independent orbital part, K_{orb}, and the T-dependent spin part, $K_s(T)$, that is proportional to the uniform susceptibility χ_s. $K_s(\text{OP})$ decreases below $T^* \sim 160$ K, followed by a rapid decrease around $T_c = 108$ K which is indicative of the bulk superconductivity at the OP, as also confirmed in the measurement of $1/T_1 T$. The behavior of $K_{ab}(T)$ suggests that the OP is almost optimally doped from a comparison with the previous study.[11] In the inset of Fig. 3, $K_{ab}(T)$ vs $K_c(T)$ plots are presented at the OP and the IP's. The spin
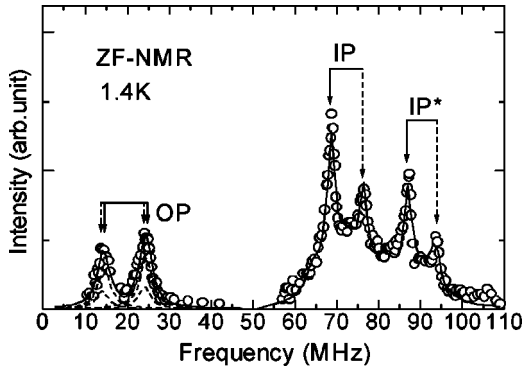
FIG. 4. A zero-field ^{63,65}Cu-NMR spectra at 1.4 K. Four peaks in $f = 55$–110 MHz, which consist of two sites (IP and IP*) and two isotopes [^{63}Cu (solid arrow) and ^{65}Cu (dashed arrow)], are observed. The spectra in $f = 10$–40 MHz correspond to the OP. The solid lines are the simulation calculated by using $^{63}\nu_Q$(IP) = 8.37 MHz, $^{63}\nu_Q$(OP) = 16.05 MHz, and H_{int} along ab plane. Each Cu moment is estimated as M(IP)$\sim 0.30\mu_B$ and M(IP*)$\sim 0.37\mu_B$ (see text). The internal field of $H_{int}\sim 0.54$ T exists even at the SC OP.

part in the measured shift $K_{s,\alpha}(T)$ at the CuO$_2$ plane is expressed as following,[17]

$$K_{s,\alpha} = (A_\alpha + 4B)\chi_{s,\alpha} \quad (\alpha = a, b, \text{ and } c \text{ axis}),$$

where A_α and B are the on-site and the supertransferred hyperfine-coupling constants. A_α is anisotropic, mainly originating from the dipole and the spin-orbit interactions for Cu-3d orbitals, and B is isotropic, originating from the Cu($3d_{x^2-y^2}$)-O($2p$)-Cu($4s$) covalent bonding. $\chi_{s,\alpha}$ is the spin susceptibility. From a linear relation in the figure, $(A_c + 4B)/(A_{ab} + 4B) \sim 0.267$ and 0.379 are estimated at the IP's and the OP, respectively. By assuming $A_c \sim -170$ kOe/μ_B and $A_{ab} \sim 37$ kOe/μ_B in YBa$_2$Cu$_3$O$_7$,[18–20] the respective values of B at the IP's and the OP in Hg-1245 are estimated as B(IP)~ 61 kOe/μ_B and B(OP) ~ 74 kOe/μ_B. These values of B are larger than the typical value of ~ 40 kOe/μ_B obtained in La$_{2-x}$Sr$_x$CuO$_4$, YBa$_2$Cu$_3$O$_7$, and YBa$_2$Cu$_4$O$_8$,[18–23] suggesting that the Cu($3d_{x^2-y^2}$)-O($2p$)-Cu($4s$) covalent bonding in Hg-1245 is stronger than in the La- or Y-based systems.

Next, we present firm evidence for the occurrence of AF ordering at the IP and the IP*. Figure 4 shows ^{63,65}Cu-NMR spectra at $H = 0$ and $T = 1.4$ K. Four and two peaks are observed in the frequency ranges of $f = 55$–110 and 10–40 MHz, respectively. The nuclear quadrupole frequencies at the IP and the OP's, ν_Q(IP) and ν_Q(OP), are estimated from the NMR experiments at high T as $^{63}\nu_Q$(IP) = 8.37 MHz and $^{63}\nu_Q$(OP) = 16.05 MHz, respectively (not shown). Therefore, all these spectra are affected by the presence of internal field H_{int} associated with the onset of AF order.

The nuclear Hamiltonian $\mathcal{H} = \mathcal{H}_Q + \mathcal{H}_Z$ at $H = 0$ below T_N is described in terms of the Zeeman interaction due to H_{int} and the nuclear electric quadrupole interaction as follows:

$$\mathcal{H}_Z = -\gamma_n \hbar \left\{ \frac{1}{2} H_\perp \cdot (I_+ + I_-) + H_\parallel \cdot I_z \right\},$$

where $H_\perp$ and $H_\parallel$ are the respective components perpendicular and parallel to the c axis and γ_n is the Cu nuclear gyromagnetic ratio, and

$$\mathcal{H}_Q = \frac{e^2 qQ}{4I(2I-1)} \{ [3I_z^2 - I(I+1)] + \eta(I_x^2 - I_y^2) \},$$

where η is the asymmetry parameter of electric-field gradient. Here, note that the quadrupole frequency $h\nu_Q \equiv 3e^2 qQ/2I(2I-1)$ and $\eta \sim 0$.

The spectra observed in $f = 55$–110 MHz correspond to the case for $\nu_Q \ll H_{int}$ due to the AF order below $T_N \sim 60$ K. Four peaks are understood as the central peaks $(1/2 \leftrightarrow -1/2$ transition) of ^{63,65}Cu at the IP and the IP*. A ratio of ^{63,65}Cu-NMR intensity at low frequency to high frequency $(I_L/I_H \sim 2)$ suggests that the two peaks at low (high) frequencies arise from the IP (IP*). The satellite peaks $(\pm 1/2 \leftrightarrow \pm 3/2$ transition) due to the electric quadrupole interaction are not well resolved. By incorporating this intensity ratio $I_L/I_H \sim 2$, $^{63}\nu_Q$(IP) = 8.37 MHz, $^{63}Q/^{65}Q \sim 1.08$, and $^{63}\gamma_n/^{65}\gamma_n \sim 0.93$, the NMR spectra at the IP and the IP* are simulated as the solid line in the figure, giving rise to the respective values of H_{int}(IP) = 6.1 T and H_{int}(IP*) = 7.7 T. These values at the IP and the IP* allow us to estimate the Cu moments M(IP)$\sim 0.30\mu_B$ and M(IP*)$\sim 0.37\mu_B$ by using a hyperfine-coupling constant $(A_{ab} - 4B) \sim -207$ kOe/μ_B where B(IP)~ 61 kOe/μ_B. These Cu moments are one-half smaller than $0.64\mu_B$ estimated in La$_2$CuO$_4$.[24] We remark that the N_h(IP) and N_h(IP*) are tentatively estimated as $[5\delta_{av} - 2N_h$(OP)$]/3 \sim 0.057 \pm 0.02$ by using an average hole content $\delta_{av} = 0.12$ evaluated from a Hall measurement.[25] Note that N_h(OP) = 0.212–0.217 was estimated via the systematic experimental relation between the N_h and the K_s at room temperature argued in the literature,[11] and hence it is shown that the OP is optimally doped. Here we assumed K_{orb} for Hg-1245 to be 0.19–0.20 from a comparison with other multilayered cuprates.[16,26]

The spectra in $f = 10$–40 MHz suggest the case for $\nu_Q \sim 16$ MHz $\simeq H_{int}$. Actually, the calculated spectra to be consistent with the experiment are indicated in the figure, allowing us to estimate $\nu_Q \sim 16$ MHz and $H_{int} = H_\perp \sim 0.54$ T. These spectra are hence assigned as arising from the OP. This H_{int} of 0.54 T is far larger than the calculated dipole field in the OP of ~ 70 Oe, which is induced by the Cu moments of M(IP)$\sim 0.30\mu_B$ and M(IP*)$\sim 0.37\mu_B$. H_{int}(OP)~ 0.54 T corresponds to the Cu moments of $\sim 0.02\mu_B$.

Figure 5 indicates the T dependence of $1/T_1 T$ of ^{63}Cu at the IP's and the OP for $H \parallel c$. Remarkably, $1/T_1 T$(OP) exhibits a pseudogap behavior below $T^* \sim 160$ K. $1/T_1$(OP) is distributed below ~ 90 K. In the normal state, a recovery curve of nuclear magnetization is consistent with a theoretical one for determining a single value of T_1 as seen in Fig. 6(a).[27] Below ~ 90 K, however, a short component in the recovery curve is observed as presented in Fig. 6(b). A tentative fitting
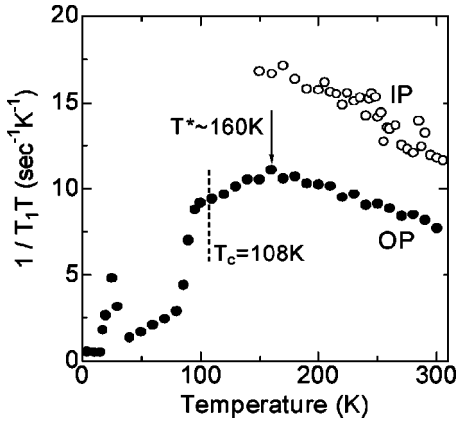
FIG. 5. T dependence of $1/T_1T$ for $H\|c$. T_1(OP) below ~90 K is the long component in the recovery curve. The pseudogap behavior is observed at the optimally doped OP below $T^*\sim160$ K. On the other hand, the underdoped IP does not show any pseudogap indication, revealing that the low-energy spectral weight in $\chi(\mathbf{q}=\mathbf{Q},\omega)$ is critically enhanced around $\omega\sim0$ toward AF ordering at $T_N\sim60$ K.

to the curve, which is indicated by a solid line, allows us to estimate a short and a long component in T_1. Their T dependencies are shown in Fig. 7, where the short components are presented by open square.

Generally in the SC mixed state under magnetic field, the short component in T_1 is believed to arise due to the presence of the *normal state* in vortex cores. But the large fraction of the short component reaching $\sim80\%$ is unusual, giving rise to almost a same fraction as the short component at $H=0$ indicated by open triangle in the figure. These results ensure that the short component dose not arise due to the presence of vortex cores, but originates from the unexpected
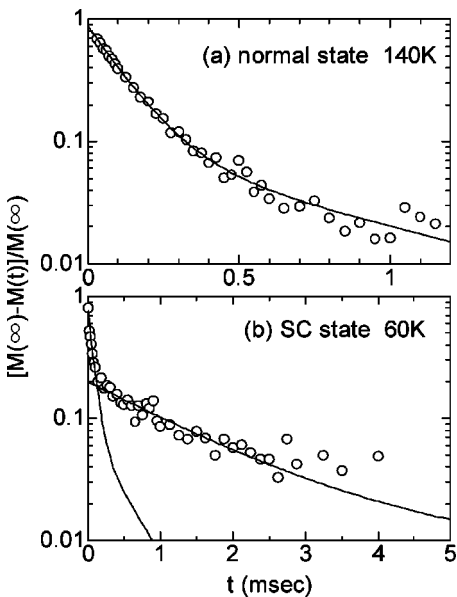

FIG. 6. The recovery curve of nuclear magnetization for determining T_1 at the OP at (a) the normal and (b) the SC state, respectively. A short component in the recovery curve is observed as presented below ~90 K in the SC state.
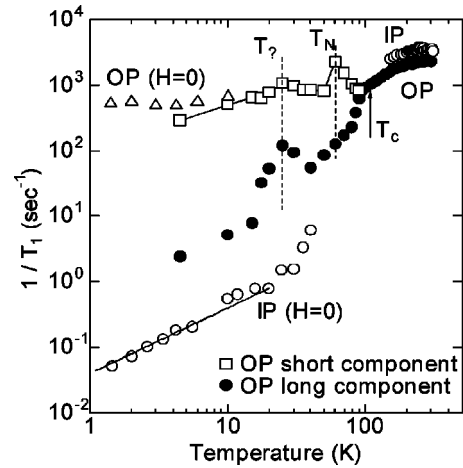

FIG. 7. T dependence of $1/T_1$ for $H\|c$. $1/T_1$ below T_N can be measured at zero field. $1/T_1$(OP) shows the peak at $T_N\sim60$ K associated with AF ordering at the IP. $1/T_1$(IP) shows a $T_1T\sim$const relation far below T_N, indicating that the IP is metallic. $[T_1(OP)/(T_1(IP)]\sim10^{-3}$ at low T shows the existence of low-lying magnetic excitations inherent to the OP, which is associated with a possible interplay with the superconductivity.

relaxation process at the SC OP. Thus, some low-lying magnetic excitations survive at the SC OP even though the d-wave superconductivity is formed well below $T_c=108$ K. The short component indicates two peaks at $T_N\sim60$ K and $T_?\sim25$ K, whereas the long component indicates a peak at $T_?\sim25$ K. $T_N\sim60$ K is indicative of an onset of AF ordering at the IP's, corroborated by the increase of K_s(OP). Recent muon spin resonance measurement also evidences an AF ordering below ~60 K in this material.[28] $T_?\sim25$ K might be related to the occurrence of H_{int}(OP) because both the long and short components show peaks.

On the other hand, $1/T_1T$(IP) increases monotonically, whereas K_s(IP) decreases with decreasing T down to 150 K. Instead of the pseudogap, unexpectedly, the NMR signals at the IP and the IP* disappear below ~150 K. This is because the low-energy spectral weight in dynamical response function $\chi(\mathbf{q}=\mathbf{Q},\omega)$ is critically enhanced around $\omega\sim0$. Here $\mathbf{Q}$ is the AF wave vector $(\pi/a,\pi/a)$. Eventually, the IP's order antiferromagnetically below T_N as evidenced from the zero-field AF NMR experiment that probes the Cu moments of M(IP)$\sim0.30\mu_B$ and M(IP*)$\sim0.37\mu_B$. The measurement of T_1 at the IP reveals a behavior of $T_1T\sim$const below ~20 K in the SC state at $H=0$ as seen in Fig. 7. In magnetically ordered metals, a nuclear-spin-relaxation process is mediated by the interaction between nuclear spins and conduction electrons via spin-wave excitations, which is called as the Weger mechanism leading to a behavior of $T_1T=$const at low T.[29,30] Thus the IP is suggested to be not in an insulating regime but a metallic one, which is consistent with the estimated hole content of $\sim0.057\pm0.02$. The small value of $T_1T=$const indicates that a large gap opens in the magnetic excitation, indicating that the AF ordering in the IP is in a static regime.

When the SC state is closely faced to the AF state realized in the doped CuO_2 plane, it is not obvious to what extent the

superconductivity is affected because of the presence of AF state. Hg-1245 is a good candidate to address this issue. Both the measurements of T_1 and Knight shift evidence that the OP is in the SC state. If the AF-spin polarization at the IP induced the $H_{int}(OP) = 0.54$ T via the hybridization between $4s(IP)$ and $4s(OP)$ and/or $3d_{3z^2-r^2}(OP)$ [not $3d_{x^2-y^2}(OP)$], a ratio of $[T_1(OP)/T_1(IP)] \sim [H_{int}(IP)/H_{int}(OP)]^2 \sim [6.1T/0.54T]^2 \sim 10^2$ would be expected. It is, however, surprising that the T_1 at the SC OP is 10^3 times shorter than at the antiferromagnetically ordered IP at low T far below T_N, being $[T_1(OP)/(T_1(IP)] \sim 10^{-3}$. This result demonstrates the existence of low-lying magnetic excitations inherent to the OP associated with a possible interplay with the superconductivity. It suggests that the weak AF order with small moment $0.02\mu_B$ is responsible for the low-lying magnetic fluctuations at the OP and coexists with the SC state at the OP. We note that this coexistence seems to be similar to the phenomenon near vortex cores where the AF correlations originating from the vortex cores extend over the cores into the SC region.[1] On the other hand, it is quite interesting issue whether the SC order parameter exists at the metallic AF IP in Hg-1245, but this is a future issue, because the NMR signal from the AF IP disappears in $T = 40$–150 K.

Finally we mentioned some results of $TlBa_2Ca_4Cu_5O_y$ (Tl-1245) with $T_c = 100$ K, which is slightly much overdoped than Hg-1245. As shown in Fig. 8(a), the $1/T_1T(OP)$ of Tl-1245 does not show the pseudogap behavior, indicating that the OP is in overdoped regime. $1/T_1T(IP)$ increases monotonically with decreasing T and the signals of the IP and the IP* disappear below ~ 140 K as well as Hg-1245. The low-lying magnetic excitations are induced at the SC OP also in Tl-1245, and thus $T_1(OP)$ distributes below ~ 60 K. Its short components show a peak at ~ 45 K, corresponding to the peak of $T_N \sim 60$ K of Hg-1245, as shown in Fig. 8(b). This $T_N \sim 45$ K at the IP of Tl-1245 suggests that the IP of Tl-1245 has somewhat much carrier content than that of Hg-1245. Interestingly, however, the second anomaly corresponding to $T_?$ disappears in Tl-1245, and the linewidth of the OP does not change between 150 K and 20 K, which is quite in contrast to Hg-1245 as shown in Figs. 8(c) and 2(b). This ensures that the internal field at the OP of Tl-1245 is quite tiny even below $T_N \sim 45$ K at the IP, which is different from $H_{int}(OP) \sim 0.54$ T of Hg-1245. These things imply that $H_{int}(OP) \sim 0.54$ T is induced below $T_? \sim 25$ K in Hg-1245.

III. SUMMARY

In summary, the $^{63,65}Cu$-NMR measurements have unraveled that the disparate electron phases emerge at the outer two CuO_2 planes and the inner three ones in $HgBa_2Ca_4Cu_5O_y$. The T dependencies of Knight shift and $1/T_1$ have revealed that the optimally doped OP undergoes the bulk SC transition at $T_c = 108$ K and the underdoped IP's
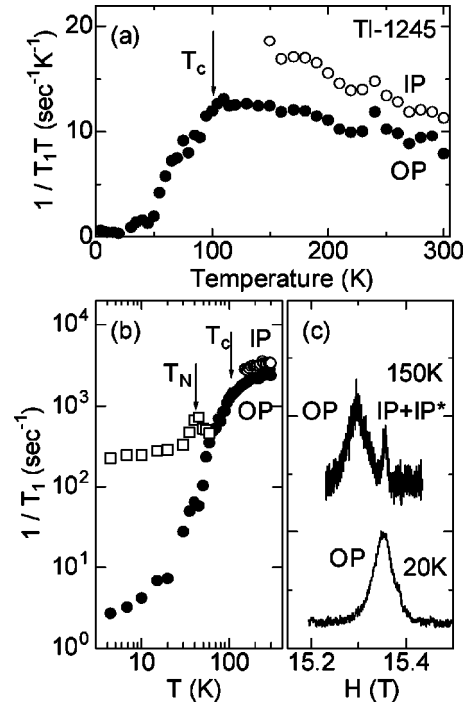

FIG. 8. T dependence of (a) $1/T_1T$ and (b) $1/T_1$ for $H\|c$ in Tl-1245, which is slightly overdoped compared with Hg-1245. The short component of $T_1(OP)$ shows the peak at ~ 45 K, indicative of AF ordering at the IP. $T_?$ is absent in Tl-1245. Figure (c) presents NMR spectra at 150 K and 20 K. No change is observed in the linewidth of the OP signal, indicating that the internal field at the OP is quite tiny, in contrast with Hg-1245.

do the AF transition below $T_N \sim 60$ K without any indication of pseudogap. The zero-field $^{63,65}Cu$-NMR experiments at low T have provided firm evidence that the respective AF moments at the IP and the IP* are $M(IP) \sim 0.30\mu_B$ and $M(IP^*) \sim 0.37\mu_B$. The bulk superconductivity with the high value of $T_c = 108$ K and the static AF ordering at $T_N = 60$ K take place even though the AF and SC layers are alternatively stacked with the respective thickness being comparable with ~ 9.7 Å and ~ 12.5 Å. The AF-spin polarization at the IP is found to induce the Cu moments of $\sim 0.02\mu_B$ at the OP that fluctuates faster than the AF moment at the IP does, evidencing the AF proximity effect into the SC OP.

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