Antiferromagnetic Vortex Core of Tl$_2$Ba$_2$CuO$_{6+\delta}$ Studied by NMR

K. Kakuyanagi$^1$, K. Kunagai$^1$, Y. Matsuda$^2$, and M. Hasegawa$^3$

$^1$Division of Physics, Graduate School of Science, Hokkaido University, Sapporo 060-0810, Japan
$^2$Institute for Solid State Physics, University of Tokyo, Kashiwanoha 5-1-5, Kashiwa, Chiba 277-8581, Japan
$^3$Institute of Material Research, Tohoku University, Katahira, Sendai, 980-8577, Japan

Spatially-resolved NMR is used to probe the magnetism in and around vortex cores of nearly optimally-doped Tl$_2$Ba$_2$CuO$_{6+\delta}$ ($T_c=85$ K). The NMR relaxation rate $T_1^{-1}$ at $^{205}$Tl site provides a direct evidence that the AF spin correlation is significantly enhanced in the vortex core region. In the core region Cu spins show a local AF ordering with moments parallel to the layers at $T_N=20$ K. Above $T_N$ the core region is in the paramagnetic state which is a reminiscence of the state above the pseudogap temperature ($T^*\simeq120$ K), indicating that the pseudogap disappears within cores.

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In high-$T_c$ cuprates (HTC) the superconductivity with $d$-wave symmetry appears when carriers are doped into the antiferromagnetic (AF) Mott insulators. It is well established that the strong AF fluctuation plays a crucial role in determining many physical properties. Therefore the relation between superconductivity and magnetism has been a central issue in the physics of HTC. Especially concerning the magnetism. The main reason is that STM experiments do not directly reflect the magnetism. Neutron scattering experiments on La$_{2-x}$Sr$_x$CuO$_4$ have reported that an applied magnetic field enhances the AF correlation in the superconducting state. However, the relation between the observed AF and the magnetism within vortex cores is not clear, because the neutron experiments lack spatial resolution. Recent $\mu$SR experiments on underdoped YBa$_2$Cu$_3$O$_{6.5}$ have reported the presence of static magnetism in vortex core region, but the detailed nature of this magnetism is still not clear.

Recent experimental and theoretical NMR studies have established that the frequency dependence of spin-lattice relaxation rate $T_1^{-1}$ in the vortex state serves as a probe for the low energy excitation spectrum which can resolve different spatial regions of the vortex lattice. Unfortunately, up to now, all of these spatially-resolved NMR measurements have been carried out at the planar $^{17}$O sites, at which the AF fluctuations are filtered due to the location of O-atoms in the middle of neighboring Cu atoms with antiparallel spins.

In this Letter we provide local information on the AF correlation in the different regions of the vortex lattice extending the measurements to the vortex core region, by performing a spatially resolved NMR imaging experiments on $^{205}$Tl-nuclei in nearly optimally-doped Tl$_2$Ba$_2$CuO$_{6+\delta}$. This attempt is particularly suitable for the above purpose because $T_1^{-1}$ at the Ti-site, $^{205}$Tl$^{-1}$, can monitor AF fluctuations sensitively. Quite generally, $1/T_1$ is expressed in terms of the dynamical susceptibility as $1/T_1 = \gamma_n k_B T \sum_{\mathbf{q}} |A_q|^2 \frac{\mu_0 \chi(q,\omega)}{\omega}$, where $\gamma_n$ is the nuclear gyromagnetic ratio, $A_q$ is the hyperfine coupling between nuclear and electronic spins, and $\omega_0$ is the Larmor frequency. Because Ti atoms are located just above the Cu atoms and there exist large transferred hyperfine interactions between Ti nuclei and Cu spin moments through apical oxygen, Ti sees the full wavelength spectrum of Cu magnetic spin fluctuation; $^{205}$Tl$^{-1}$ is dominated by $\chi(q)$ at $q=(\pi,\pi)$, i.e. AF fluctuations. This should be contrasted to the O-sites at which $\chi(q)$ is dominated by uniform fluctuations at $q=(0,0)$.

NMR measurements were carried out on the $c$-axis oriented polycrystalline powder of high quality Tl$_2$Ba$_2$CuO$_{6+\delta}$ ($T_c=85$ K) in the external field ($H_0=2.1$ T) along the $c$-axis. The $^{205}$Tl spin echo sig-
nals were obtained by a pulse NMR spectrometer. The spectra was obtained by convolution of the respective Fourier-transform-spectra of the spin echo signals measured with an increment of 50 kHz. A very sharp spectrum (∼50 kHz) above \( T_c \) becomes broad below \( T_c \) due to the development of vortices. The solid lines in Fig. 1 and lower inset depict the NMR spectra at 5 K and 20 K, respectively. A clear asymmetric pattern of the NMR spectra was obtained by a pulse NMR spectrometer. The real spectrum broadens due to the imperfect orientation of the power and distortion of the vortex lattice. The red dotted lines in Fig. 1 and lower inset represent the simulation spectra in which the Lorentzian broadening function, \( f(\omega_{loc}) = \sigma / (4\hbar^2 \omega_{loc}^2 + \sigma^2) \), is convoluted to \( \omega_{loc} \) at \( T_0 = 20 \text{K} \) (inset). On the other hand, the spectrum at 5 K shows significant broadening at high frequency region (core region), while it can be well fitted below 52.1 MHz (main panel). The filled circles in Fig. 2 display the line width at the high frequency tail, \( \Delta f \), which is defined as a difference between the frequency at the peak intensity and the frequency at which the intensity becomes 1% of the peak. For the comparison, the line width calculated from the simulation spectra without taking into account the core magnetism. Open circles represent \( \delta f \) which will be discussed later, the histogram shows the low and high frequency cutoffs at the center of the vortex lattice (C-point) and at the center of cores (A-point), respectively, and shows a peak at the field corresponding to the saddle point (B-point). This characteristic spectrum (Redfield pattern) demonstrates that the NMR frequency depends on the position of the vortex lattice. We therefore can obtain the spatially-resolved information of the low energy excitation by analyzing the frequency distribution of the corresponding NMR spectrum.

We first discuss the observed NMR spectra. The real spectrum broadens due to the imperfect orientation of the power and distortion of the vortex lattice. We first discuss the observed NMR spectra. The real spectrum broadens due to the imperfect orientation of the power and distortion of the vortex lattice. The red dotted lines in Fig. 1 and lower inset represent the simulation spectra in which the Lorentzian broadening function, \( f(\omega_{loc}) = \sigma / (4\hbar^2 \omega_{loc}^2 + \sigma^2) \), is convoluted to \( \omega_{loc} \). The theoretical curve reproduces the data well in the whole frequency range at \( T=20 \text{K} \) (inset). On the other hand, the spectrum at 5 K shows significant broadening at high frequency region (core region), while it can be well fitted below 52.1 MHz (main panel). The filled circles in Fig. 2 display the line width at the high frequency tail, \( \Delta f \), which is defined as a difference between the frequency at the peak intensity and the frequency at which the intensity becomes 1% of the peak. For the comparison, the line width calculated from the simulation spectra with Lorentzian broadening function is plotted by the dashed line. At high temperature \( \Delta f \) agrees well with the calculation, while below 20 K it becomes much larger. We also plot \( \delta f \) which represents the line width at the half intensity (open circles). The fact that \( \delta f \) changes little below 20 K confirms that the line broadening occurs only in the high frequency core region. In \( \mu \)SR experiments, the high frequency tail was
attributed to the static magnetism around cores, which causes additional broadening [10]. It should be noted that because of large transfer hyperfine coupling between Tl nuclei and Cu moment, the broadening of Tl-NMR spectrum associated with the static magnetism is more pronounced than that of $\mu$SR spectrum. Therefore the observed broadening below $\sim 20$ K is naturally explained by the appearance of static magnetism within cores below $\sim 20$ K. To obtain deep insight into this phenomena, the measurements of $T_1$ are crucial.

For ${}^{205}\text{Tl}$ with nuclear spin $I=1/2$, the recovery curve of the nuclear magnetization $M(t)$ fits well to a single exponential relation, $R(t) = (M(\infty) - M(t))/M(\infty) = \exp(-t/T_1)$ in the normal state. In the vortex state, on the other hand, the feature of the recovery curves is strongly position dependent as shown in Fig. 3. The spin echo intensities are measured as a function of $\tau$ after saturation pulses. Then the nuclear magnetization recovery curves are obtained from each frequency component of the Fourier transform spectra. We obtained the data set of the recovery intensity for each frequency point at the 28 kHz interval with a gaussian weight function of $\sigma = 10$ kHz. There are two distinct features. First, the decay time at cores is much faster than at saddle points. Second, while the recovery curves show the single exponential at the saddle point, they show a $\sqrt{\tau}$ dependence at the core region as shown in the inset of Fig. 3. We will discuss this $\sqrt{\tau}$ dependence later. In what follows, we define $T_1$ as the time required for the nuclear magnetization to decay by a factor $1/e$, in order to define $T_1$ uniquely for either decay curve.

The red filled circles in Fig. 1 show the frequency dependence of $205T_1^{-1}$. On scanning from outside into cores, $205T_1^{-1}$ increases rapidly after showing a minimum near saddle point. The magnitude of $205T_1^{-1}$ in the core region is almost two orders of magnitude larger than that near saddle point. This large enhancement of $205T_1^{-1}$ is in striking contrast to $17T_1^{-1}$ at $^{17}$O sites reported in YBa$_2$Cu$_3$O$_7$ [11, 12] and YBa$_2$Cu$_4$O$_8$ [13], in which the enhancement of $17T_1^{-1}$ at the core region is 2-3 times at most and has been attributed to LDOS produced by a Doppler shift of the QP energy spectrum by supercurrents around the vortices [17]. It should be noted that the LDOS effect is absent in $205T_1^{-1}$, because there are no conduction electrons at $^{205}$Tl-site. Therefore, the remarkable enhancement of $205T_1^{-1}$ provides a direct evidence that the AF correlation is strongly enhanced near the vortex core region [18]. The decrease of $205T_1^{-1}$ well outside the core when going from point C to B was also reported in $17T_1^{-1}$ [13, 14].

Figures 4 (a) and (b) depict the $T$-dependences of $(205T_1T)^{-1}$ and $205T_1^{-1}$ within cores (filled circles) and at the frequency corresponding to saddle points (open circles), respectively. In these figures, $205T_1^{-1}$ within cores, $(205T_1^{-1})_{\text{core}}$, were determined with using integrated intensities over the high frequency region beyond the A point. From high temperatures down to about 120 K, $205T_1^{-1}$ obeys the Curie-Weiss law, $(205T_1T)^{-1} \propto 1/(T + \theta)$. The lowest $T$ at which this law holds is conveniently called the pseudogap temperature $T^*$. Below $T^*$, $(205T_1T)^{-1}$ decreases rapidly without showing any anomaly associated with the superconducting transition at $T_c$, similar to other HTC [12].

The $T$-dependence of $(205T_1^{-1})_{\text{core}}$ contains some key features for understanding the core magnetism. The first important signature is that $1/(205T_1^{-1})_{\text{core}}$ exhibits a sharp peak at $T=20$ K, which we label as $T_N$ for future reference (Fig. 4(a)). Below $T_N$ $(205T_1^{-1})_{\text{core}}$ decreases.
rapidly with decreasing $T$. There are two possible origins for this peak. One is the reappearance of the pseudogap and the other is the occurrence of a local static AF (or SDW) ordering in the core region. Generally $T_{1}^{-1}$ shows a sharp peak when the AF ordering occurs. The fact that the sharp peak of $(205T_{1}^{core})^{-1}$ is observed at $T_N$ while $1/T_1$ at the saddle points shows neither a peak nor broad maximum at $T^*$ as shown in the inset of Fig. 4(b) excludes the possibility of pseudogap, supporting the AF ordering. Moreover, as shown in Fig. 2, broadening of the NMR spectrum starts at $\sim$20 K, which coincides with $T_N$. This fact gives an additional strong evidence on AF ordering. We also point out that the appearance of the local AF ordering is also consistent with the $\sqrt{T}$ dependent nuclear magnetization decay curve shown in the inset of Fig. 3. In fact, the $\sqrt{T}$ dependence has been observed when the microscopic inhomogeneous distribution of $T_{1}^{-1}$ due to strong magnetic scattering centers is present [20]. On basis of these results, we are lead to conclude that the local AF ordering takes place in the core region at $T_N=20$ K; $T_N$ corresponds to the Néel temperature within the core. This AF ordering is consistent with the prediction of recent theories based on the $t-J$ and SO(5) models [6]. The present results also should be distinguished from those of the neutron scattering experiments [1] and 2). (2) Upon approaching the vortex core, $(205T_{1}^{core})^{-1}$ is strongly enhanced (Fig. 1). (3) Near the core region, the NMR recovery curves show the $\sqrt{T}$-dependence (Fig. 3). (4) $(205T_{1}^{core})^{-1}$ exhibits a sharp peak at $T=20$ K (Fig. 4). All of these results provide direct evidence that in the vortex core region the AF spin correlation is extremely enhanced, and that the paramagnetic-AF ordering transition of the Cu spins takes place at $T_N=20$ K. We also find the pseudogap disappears within the core. The present results offer a new perspective on how the AF vortex core competes with the $d$-wave superconductivity.

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