NMR studies on the internal structure of high-T_c superconductors and other anorganic compounds

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(Dated: March 31, 2006)

Spatially-resolved NMR is used to probe internal structures in highly correlated superconductors of optimally-doped $Tl_2Ba_2CuO_{6+\delta}$ (T_c=85 K) and a heavy fermion superconductor $CeCoIn_5$ (T_c=2.3 K). The characteristic change of the properties of ^{205}Tl-NMR in the vortex state provides a clear evidence of the antiferromagnetic order in the vortex cores below 20 K in $Tl_2Ba_2CuO_{6+\delta}$. We also obtain anomalous ^{115}In-NMR spectra of $CeCoIn_5$, which provides a microscopic evidence for the occurrence of a spatially-modulated superconducting order parameter expected in a Fulde-Ferrel-Larkin-Ovchinnkov (FFLO) state.

Keywords: NMR, superconductivity, vortex core, antiferromagnetic order, FFLO state
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INTRODUCTION

The relation between superconductivity (SC) and magnetism has been recognized as a central issue in the physics of highly correlated materials such as high-T_c cuprates (HTSC) and heavy fermion compounds. The microscopic structure of the vortex state in those substances comes out to be a very interesting subject. [1] Recently, the coexistence or competition of SC and magnetic ground states have been intensively studied [2, 3]

Very recent heat capacity measurements of heavy fermion compound $CeCoIn_5$ revealed that a second order phase transition takes place at $T^{\#}(H)$ within the SC state in the vicinity of the upper critical field at low temperatures [4, 5]. The transition line branches from $H_{c2}^{\parallel}$-line and decreases with decreasing T, indicating the presence of a novel SC phase. While recent experimental results make the Fulde-Ferrel-Larkin-Ovchinikov (FFLO) [6, 7] scenario a very appealing one for $CeCoIn_5$, there is no direct experimental evidence so far which verifies the spatially nonuniform SC state expected in the FFLO state.

As NMR is particularly suitable for the above purpose because NMR can monitor the local information about low energy quasiparticle excitations and antiferromagnetic fluctuations sensitively. In this paper, we report new findings associated with the microscopic vortex structure, namely, antiferromagnetic vortex core in an optimally-doped HTSC, $Tl_2Ba_2CuO_{6+\delta}$, [8] and the spatially modulated superconducting gap (FFLO) state in a strong magnetic field exceed a Pauli limit in $CeCoIn_5$.[9]

EXPERIMENTAL

NMR measurements were carried out on the c-axis oriented polycrystalline powder of $Tl_2Ba_2CuO_{6+\delta}$ (T_c=85 K) and also on a single crystal of $CeCoIn_5$ in the magnetic field $\boldsymbol{H}$ parallel to the [100]-direction. The nuclear spin-lattice relaxation time of ^{205}Tl in $Tl_2Ba_2CuO_{6+\delta}$ and the Knight shift of ^{115}In-NMR at the In(1) site with axial symmetry were intensively investigated.

RESULTS AND DISCUSSION

A clear asymmetric pattern of the ^{205}Tl-NMR spectrum, which originates from the local field distribution associated with the vortex lattice (the Redfield pattern), is observed below the vortex lattice melting temperature (∼60 K at H=2.1 T). The solid line in Fig. 1. depicts the NMR spectra at T=5 K and H=2.1 T. The local field profile in the vortex state shown in the inset of Fig. 1 is given by approximating $H_{loc}(\boldsymbol{r})$ with the London result for square vortex lattice. The thin solid line in Fig. 1 depicts the histogram at a particular local field which is given by the local field distribution $f(H_{loc}) = \int \delta[H_{loc}(\boldsymbol{r}) - H_{loc}]d^2\boldsymbol{r}$. In the calculation we used ξ_{ab}=18Å and $\lambda_{ab} = 1700$ Å, and assumed the square vortex lattice. The theoretical curve reproduces the data well above T=20 K. On the other hand, the spectrum at T=5 K shows significant broadening at high frequency region (core region), while it can be well fitted below 52.1 MHz as seen in the inset of Fig. 1. The line broadening occurs only in the high frequency core region. The observed broadening below ∼20 K is explained by the appearance of static magnetism within vortex cores the below ~20 K

The filled circles in Fig. 1 show the frequency dependence of $^{205}T_1^{-1}$. On scanning from outside into cores, $^{205}T_1^{-1}$ increases rapidly after showing a minimum near saddle points. The magnitude of $^{205}T_1^{-1}$ in the core region is almost two orders of magnitude larger than that near the saddle point. The remarkable enhancement of $^{205}T_1^{-1}$ provides a direct evidence that the AF correlation is strongly enhanced near the vortex core region. Figures 2 (a) and (b) depict the T-dependences of $(^{205}T_1T)^{-1}$ and $^{205}T_1^{-1}$ within cores (filled circles) and at the saddle points (open circles), respectively. From high temperatures down to about 120 K, $(^{205}T_1T)^{-1}$ obeys the Curie-Weiss law, $(^{205}T_1T)^{-1} \propto 1/(T+\theta)$. The lowest T at which this law holds is conveniently called the pseudogap temperature T^*. Below T^*, $(^{205}T_1T)^{-1}$ decreases rapidly without showing any anomaly associated with the superconducting transition at T_c, similar to other HTC. The important signature is that the T-dependences of T_1^{-1} in the vortex core have a peak around T=20 K, and below T ~20K, $(^{205}T_1^{core})^{-1}$ decreases rapidly with decreasing T as seen in Fig. 2 (b).

On basis of the results of the broadening of the NMR spectra below 20K and the T-dependence of $1/T_1$, 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. [8] The experimental feature is well explained with the model of the AF vortex core. This AF ordering is consistent with the prediction of recent theories based on the SO(5) and $t-J$ models [2, 3].

Figure 3 shows ^{115}In-NMR spectra of $CeCoIn_5$ at various temperatures down to 120 mK at H=11.8 T (in the normal state) and H=11.3 T (across the normal to the SC phase at T~700 mK and also the second phase transition at T~300 mK)). The spectrum with single peak is observed down to $T^{\#}$(=300 mK) and the peak position is slightly decrease with decreasing temperature. Then, the spectrum shows complex feature just across the boundary of the high field SC phase at $T^{\#}$. Below $T^{\#}$, the spectrum shows double peaks. The separation of the upper and lower peak position increases with decreasing temperature. The relative ratio of the intensities at each peak changes drastically.

The Knight shift at H=11.3 T shows quite unusual temperature dependence as seen in Fig. 4. Here, the Knight shift is evaluated from the peak position by taking into account the shift due to large electric quadrupole interaction with using a second order perturbation calculation for parameters, ν_Q =8.173 MHz and η=0 for the ^{115}In(1) site (nuclear spin: I=9/2). Just across the high field SC phase at $T^{\#}$, the Knight shift of the upper one increases largely with lowing temperature. The Knight shift of the upper peak is coincident with the values in the normal state at the lowest temperature.

In the SC state, the penetration depth for NMR rf excitation field, H_{rf}, is reduced largely. In the FFLO state, the SC order parameter is nearly sinusoidal along the vortex direction (parallel to H_0). With the nodal sheet structure perpendicular to H_0 (along the a-axis), the penetration depth of H_1 is expected to be modulated. For following discussions about a simulation of the spectrum, we use a simple sinusoidal relation of the FFLO SC gap with the relation of

$$\Delta(x) = \Delta_0 \sin Qx$$

near H_{C2}. [10] From the London equation, the spatial distribution of H_{rf} in the FFLO state is given by

$$H_1(x) \propto \frac{\sinh\frac{\Lambda/2-x}{\lambda} + \sinh\frac{x}{\lambda}}{\sinh\frac{\Lambda}{2\lambda}}$$

at the boundary condition that H_1 is equal to that of the normal state at $x=0$ and $x=\Lambda/2$. With using the spatial distribution of H_1 amplitude, the NMR spectrum is given by

$$I(k) = \int_0^{\Lambda/2} \delta\left(K\left(\frac{\Delta(x)}{T}\right) - k\right)[H_1(x)]^3 \,\mathrm{d}x$$

Here, $K\left(\frac{\Delta(x)}{T}\right)$ is the Yoshida function for the Knight shift in the SC state. Here, adjustable parameters are only two, $\frac{\Lambda}{\lambda}$ and $\frac{T}{\Delta_0}$.

Our simulations seem to interpret very well the split spectrum with reasonable change of the Knight shift and the intensity as shown in Fig. 5. The important result seen from the simulation analysis is that the wave length of the spatial oscillation of the SC order parameter in the FFLO state decreases with lowering temperature. Near the transition boundary between non-modulated SC and the FFLO phase at $T^{\#}$=300 mK, Λ is 50 times larger than the penetration length, $\lambda \sim 2000$ Å just below $T^{\#}$. At temperature where Λ/λ, is comparable with $10 \sim 1$, the NMR line shape changes drastically. At low temperature where Λ is smaller than λ, the NMR intensity from the SC region becomes dominant and is temperature-independent. The simulations in good agreement with the experimental spectra indicats the existence of the spatially modulated SC order gap as expected in the FFLO phase.

In summary, from the spatially-resolved ^{205}Tl-NMR in $Tl_2Ba_2CuO_{6+\delta}$. and ^{115}In(1)-NMR in $CeCoIn_5$, we have obtained novel features for the internal structure of superconductors associated with the vortex state. In the vortex core region in $Tl_2Ba_2CuO_{6+\delta}$, the AF spin correlations are extremely enhanced, and that the paramagnetic-AF order transition of the Cu moments takes place at T_N ~20 K. The anomalous change of the ^{115}In(1)-NMR spectra in $CeCoIn_5$ are well characterized by taking into account a spatial modulation of the SC gap and the penetration depth of rf field (H_1). The present study provide the strong evidence from a microscopic point of view that the high field phase with in SC state in $CeCoIn_5$ is of the FFLO phase.

[1] S.A. Kivelson *et al.*, Rev. Mod. Phys. **75** 1201 (2003).
[2] S.C. Zhang, Science **275** (1997) 1089.
[3] A. Himeda *et al.*, J. Phys. Soc. Jpn. **66** (1997) 3367, M. Ogata, Int. J. Mod. Phys. **B 13** (1999) 3560.
[4] H.A. Radovan *et al.*,Nature **425** (2003) 51.
[5] A. Bianchi *et al.*, Phys. Rev. Lett. **91** (2003) 187004.
[6] P. Fulde and R.A. Ferrel, Phys. Rev. A **135** (1964) 550.
[7] A.I. Larkin and Y.N. Ovchinnikov, Sov. Phys. JETP **20** (1965) 762.
[8] K. Kakuyanagi, K. Kumagai, Y. Matsuda, M. Hasegawa, Phys. Rev. Lett. **90** (2002) 197003.
[9] K. Kakuyanagi *et al.*, Phys. Rev. Lett. (2004) .
[10] M. Tachiki *et al.*, Z. Phys. B **100** (1996) 369.

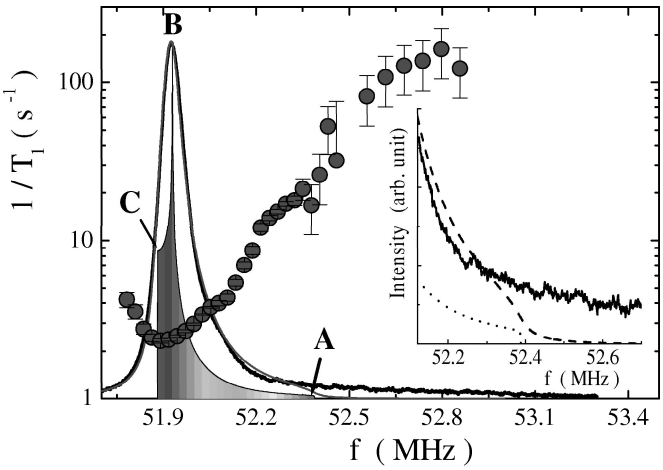

FIG. 1: ^{205}Tl-NMR spectrum (solid line) at 5 K. The intensity is plotted in a linear scale. The thin solid line depict the histogram at particular local fields of the Readfield pattern. The dotted line represents the spectrum convoluted with Lorentzian broadening function, $f(H_{loc}) = \sigma/(4H_{loc}^2 + \sigma^2)$ using σ=42kHz. The filled circles show the frequency dependence of $^{205}T_1^{-1}$ at the Tl site. The inset shows detailed one around vortex core in the enlarged scale. A , B and C represnt the poistion of center of vortex core, saddle point and center of vortex lattice for the field distribution in the square vortex lattice, respectively.

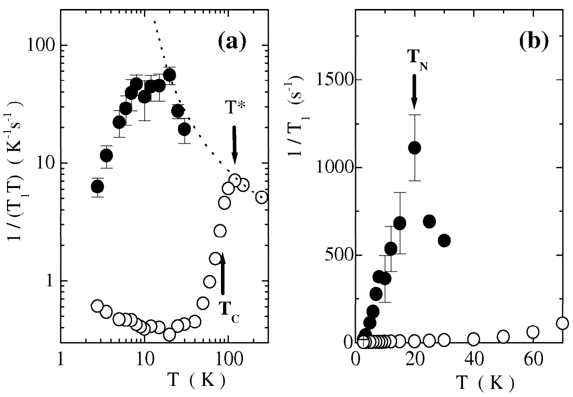

FIG. 2: T-dependence of $(^{205}T_1T)^{-1}$ **(a)** and of $^{205}T_1^{-1}$ **(b)** at low temperatures. The filled and open circles represent the data at vortex cores and at the saddle point, respectively. $T^* \simeq 120$ K is the pseudogap temperature. The dotted line represents the Curie-Weiss law which is determined above T^*. In (b), T_N is the temperature at which $^{205}T_1^{-1}$ at the core exhibits a peak.

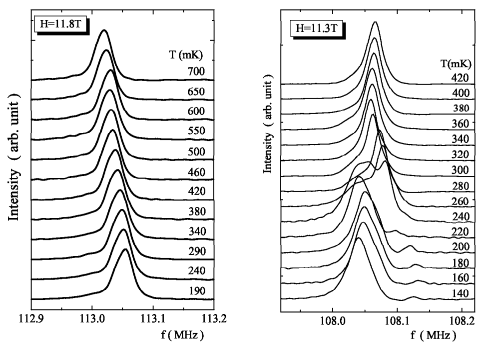

FIG. 3: ^{115}In-NMR spectra (central line of ($\pm 1/2 \leftrightarrow \mp 1/2$ transition) in $CeCoIn_5$ as a function of frequency for various temperatures at H=11.3 T.

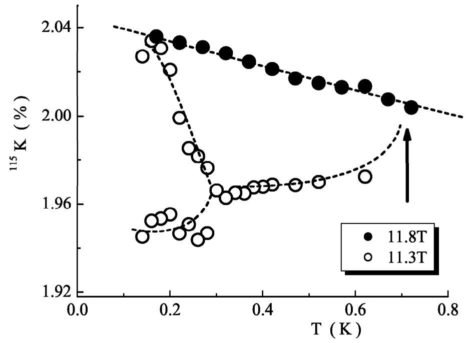

FIG. 4: Temperature dependence of Knight shift ^{115}K at H=11.8 T (•) and 11.3 T (○) in $CeCoIn_5$. The arrow shows the SC transition temperature at corrsponding field. The broken lines are guide for eyes.

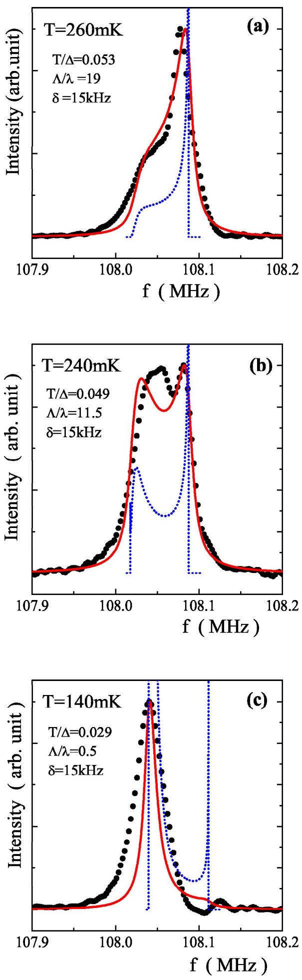

FIG. 5: ^{115}In-NMR spectra (•) at T=260 mK, T=240 mK and T=140 mK . The solid line and the dotted line represent the simulation spectra with and without a convolution of an inhomogeneous broadening with a Lorentian function (σ=15 kHz), respectively.