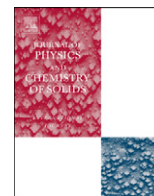




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## Novel superconducting phases in copper oxides and iron-oxypnictides: NMR studies

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## ABSTRACT

We reexamine the novel phase diagrams of antiferromagnetism (AFM) and high- $T_c$  superconductivity (HTSC) for a disorder-free  $\text{CuO}_2$  plane based on an evaluation of local hole density ( $p$ ) by site-selective Cu-NMR studies on multilayered copper oxides. Multilayered systems provide us with the opportunity to research the characteristics of the disorder-free  $\text{CuO}_2$  plane. The site-selective NMR is the best and the only tool used to extract layer-dependent characteristics. Consequently, we have concluded that the uniform mixing of AFM and SC is a general property inherent to a single  $\text{CuO}_2$  plane in an underdoped regime of HTSC. The  $T=0$  phase diagram of AFM constructed here is in quantitative agreement with the theories in a strong correlation regime which is unchanged even with mobile holes. This *Mott physics* plays a vital role for mediating the Cooper pairs to make  $T_c$  of HTSC very high. By contrast, we address from extensive NMR studies on electron-doped iron-oxypnictides  $\text{La}1111$  compounds that the increase in  $T_c$  is not due to the development of AFM spin fluctuations, but because the structural parameters, such as the bond angle  $\alpha$  of the  $\text{FeAs}_4$  tetrahedron and the  $a$ -axis length, approach each optimum value. Based on these results, we propose that a stronger correlation in HTSC than in FeAs-based superconductors may make  $T_c$  higher significantly.

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### 1. Introduction

Despite more than 24 years of intensive research, the origin of high-temperature copper-oxide superconductivity (HTSC) has not yet been well understood. HTSC can be observed on a  $\text{CuO}_2$  plane when an antiferromagnetic Mott insulator is doped with mobile carriers. A strong relationship between antiferromagnetism (AFM) and superconductivity (SC) is believed to be the key to understanding the origin of remarkably high SC transition temperatures [1–19]. As a matter of fact, site-selective  $^{63}\text{Cu}$ -NMR studies on multilayered cuprates revealed that AFM uniformly coexists with SC in a single  $\text{CuO}_2$  plane [20–25]; Square-type inner  $\text{CuO}_2$  planes (IPs) are located so far from the charge reservoir layers (CRLs), in which the disorder is introduced by the chemical substitution, exhibiting homogeneous hole doping. In the previous papers, the NMR investigations of five-layered ( $n=5$ ) compounds [20,22,25] and four-layered ( $n=4$ ) compounds [21,23,24] have revealed that the AFM quantum critical point (QCP) takes place around the hole densities  $p_c \sim 0.17$  and  $\sim 0.15$ , respectively. These  $p_c$ 's are significantly larger than  $p_c \sim 0.02$  for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  ( $n=1$ :LSCO) [26] and  $p_c \sim 0.55$  for

$\text{YBa}_2\text{Cu}_3\text{O}_{6+y}$  ( $n=2$ :YBCO) [27]. Furthermore,  $p_{max} \sim 0.22$  giving rise to the maximum of  $T_c$  for the  $n=4$  and 5 compounds is also larger than  $p_{max} \sim 0.16$  for the  $n=2$  compounds.

Upon doping with holes or electrons—e.g., by replacing some of the out-of-plane atoms with different oxidation states—AFM is destroyed and the materials become superconductors at lower temperatures. For example, in LSCO,  $\text{La}^{3+}$  is exchanged with  $\text{Sr}^{2+}$ , which increases the planar  $\text{CuO}_2$  hole density. The situation with the in-plane hole density and distribution in other superconducting cuprates is even less clear. Indirect chemical methods like solid solutions [28], semiempirical bond valence sums determined from structural bond lengths [29–31] or methods based on the Fermi surface topology [32] are used to determine the hole density. To the best of our knowledge there are no direct physical measurements that can establish a phase diagram in terms of the planar  $\text{CuO}_2$  hole density.

Under this situation, using NMR/NQR (nuclear quadrupole resonance) that can distinguish between the various atoms in the unit cell, Zheng et al. had attempted to interpret the nuclear quadrupole interaction for planar Cu and O, in terms of the local hole densities (see, e.g., Ref. [33] and references cited in it). This is indeed a sensible approach since the electric field gradient (EFG) at a nuclear site is very sensitive to changes in the local charge distribution, namely the planar Cu and O EFGs show a pronounced doping dependence. In these analyses, Zheng et al. used the

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experimental data available until 1995 with the help of atomic and cluster calculations to connect the hole densities to the EFG tensors for Cu and O. Furthermore, in order to estimate each hole density at IPs and pyramidal outer planes (OPs) in multilayered compounds, we have used an empirical relationship between the spin component in Knight shift  $K_s^{\text{ab}}(\text{RT})$  at room temperature and the planar  $\text{CuO}_2$  hole density ( $p$ ) determined with NMR/NQR.

Meanwhile, Hasse et al. [34] performed an almost model independent analysis to relate the hole densities to the EFG tensors without relying on calculations and with the extensive experimental data until 2004 including those for the parent compounds of LSCO and YBCO. On the basis of this more precise analysis to relate the hole densities to the EFG tensors than before [33], Shimizu et al. [35] have constructed a phase diagram of the  $n=2$  apical-fluorine compounds  $\text{Ba}_2\text{CaCu}_2\text{O}_{4(\text{F}, \text{O})_2}(\text{O}212\text{F})$  using a renewed empirical relationship of  $K_s(\text{RT})$  versus  $p$ . Remarkably, thus obtained phase diagram has coincided with the well established ones for the  $n=2$  YBCO and  $\text{Bi}_2\text{Ba}_2\text{CaCu}_2\text{O}_{8+y}$  ( $\text{Bi}2212$ ), exhibiting the maximum of  $T_c$  at  $p_{\text{max}} \sim 0.16$ . Thus, the universal phase diagram has been established on the  $n=2$  compounds independent of the methods to estimate the hole densities. Motivated by this fact, in the first part of the paper, we reexamine the phase diagrams of AFM and SC in multilayered compounds reported extensively in the literatures [20–24].

In the second part, we review extensive NMR studies on electron-doped iron-oxypnictides La1111 compounds in order to shed light on novel normal-state properties and their relevance with unique SC characteristics and address remarkable differences from those in HTSC.

## 2. High- $T_c$ cuprates

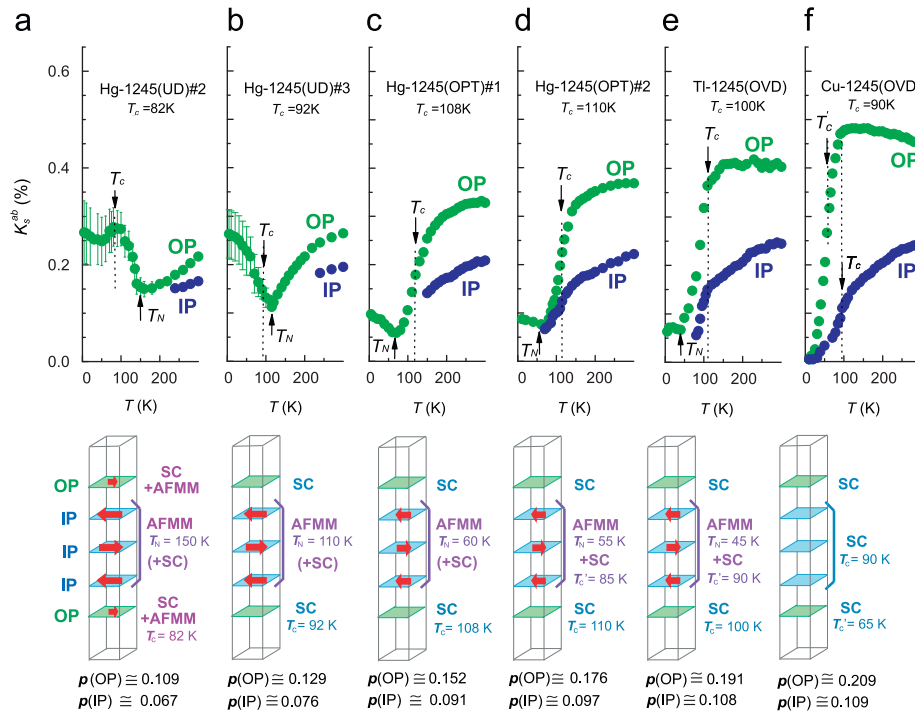
### 2.1. NMR results on five-layered ( $n=5$ ) compounds

Fig. 1 shows temperature ( $T$ ) dependence of the spin component of Knight shift  $K_s^{\text{ab}}(T)$  ( $\perp$  the  $c$ -axis) for  $n=5$  compounds such as (a)

Hg-1245(UD)#2 ( $T_c=82$  K) [25], (b) Hg-1245(UD)#3 ( $T_c=92$  K) [25], (c) Hg-1245(OPT)#1 ( $T_c=108$  K) [36], (d) Hg-1245(OPT)#2 ( $T_c=110$  K) [22], (e) Tl-1245(OVD) ( $T_c=100$  K) [22,36], and (f) Cu-1245(OVD) ( $T_c=90$  K) [37].  $K_s^{\text{ab}}(\text{RT})$  at room temperature is proportional to the planar  $\text{CuO}_2$  hole density  $p$  [33,37]. Here,  $p$  for IPs and OPs is independently estimated using the renewed relationship of  $p$  versus  $K_s(\text{RT})$ ,  $p=0.502K_s(\text{RT})-0.014$  [35].

In the  $T$  dependence of  $K_s^{\text{ab}}(T)(\text{OP})$ , a unusual upturn was observed below temperatures marked by up-arrows in Figs. 1(a)–(e), below which the linewidth of  $^{63}\text{Cu}$ -NMR spectra at OPs steeply increases. Note that this upturn in  $K_s^{\text{ab}}(T)$  at OPs was associated with an onset of AFM order at IPs, which was also corroborated by a critical enhancement of  $1/T_1$  in the previous studies on the other Hg-1245 compounds [22,36]. As a result of reduction of hole densities, it was demonstrated that the Néel temperature  $T_N$  at IPs increases from  $T_N=45$  K for (e) Tl-1245(OVD) ( $T_c=100$  K) [22,36] to  $T_N=150$  K for (a) Hg-1245(UD)#2 ( $T_c=82$  K) [25]. Layer-dependent physical properties are summarized in lower panels of Figs. 1(a)–(f).

Spontaneous AFM moments  $M_{\text{AFM}}(\text{IP})$  at IPs and OPs for the  $n=5$  compounds were determined with the zero-field Cu-NMR at 1.5 K. This is because an internal field ( $H_{\text{int}}$ ) is induced by  $M_{\text{AFM}}(\text{IP})$  at the Cu sites in IPs and OPs.  $M_{\text{AFM}}(\text{IP})$  is evaluated by using the relation  $H_{\text{int}}(\text{IP})=|A_{\text{hf}}(\text{IP})|M_{\text{AFM}}(\text{IP})$  with the hyperfine coupling constant  $A_{\text{hf}}(\text{IP})=-20.7T/\mu_B$  [36]. The details are referred to the literatures [22,25,36]. The values of  $M_{\text{AFM}}(\text{IP})$  ranging in  $0.04-0.20\mu_B$  are significantly reduced by the mobile holes from  $0.5-0.7\mu_B$  in undoped cuprates [39,40]. This is consistent with the conclusion of neutron diffraction measurement on (a) Hg-1245(UD)#2 ( $T_c=82$  K) [25]; their magnetic moments were suggested to be less than  $0.3\mu_B$  if any, since a Bragg intensity, that enables to estimate  $M_{\text{AFM}}$ , was not apparent [41]. The NMR outcome on (a) Hg-1245(UD)#2 ( $T_c=82$  K) ensures that the SC with  $T_c=82$  K uniformly coexists with the AFM order of  $M_{\text{AFM}}(\text{OP}) \sim 0.05\mu_B$  at OPs [25]. This result convinces along with the previously reported ones that the uniform coexistence of AFM and SC is a general event



**Fig. 1.** Temperature dependence of  $K_s^{\text{ab}}$  for (a) Hg-1245(UD)#2 ( $T_c=82$  K) [25], (b) Hg-1245(UD)#3 ( $T_c=92$  K) [25], (c) Hg-1245(OPT)#1 ( $T_c=108$  K) [36], (d) Hg-1245(OPT)#2 ( $T_c=110$  K) [22], (e) Tl-1245(OVD) ( $T_c=100$  K) [22,36], and (f) Cu-1245(OVD) ( $T_c=90$  K) [37]. The hole density  $p$  for IPs and OPs is independently estimated using the renewed relationship of  $p$  versus  $K_s(\text{RT})$ ,  $p=0.502K_s(\text{RT})-0.014$  [35] (see the text). Layer-dependent physical properties are summarized in lower panels.

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