Spin pseudo–gap and interplane coupling in $Y_2Ba_4Cu_7O_{15}$: a $^{63}$Cu nuclear spin–spin relaxation study

R. Stern, M. Mali, J. Roos and D. Brinkmann
Physik-Institut, Universität Zürich, CH–8057 Zürich, Switzerland
(October 28, 1994)

We report measurements of the Gaussian contribution, $T_{2G}$, to the plane $^{63}$Cu nuclear spin–spin relaxation time in the $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$ blocks of normal and superconducting $Y_2Ba_4Cu_7O_{15}$. The data confirm our previous results that adjacent CuO$_2$ planes have different doping levels and that these planes are strongly coupled. – The static spin susceptibility at the antiferromagnetic wave vector exhibits a Curie–Weiss like temperature dependence in the normal state.

The $Y_2Ba_4Cu_7O_{15}$ data are incompatible with a phase diagram based on a single CuO$_2$ plane theory and suggest that the appearance of a spin gap implies interplane coupling. Additional data for $YBa_2Cu_3O_8$ and $YBa_2Cu_4O_{6.982}$ are in accord with the single plane theory. – The temperature dependence of $T_{2G, ind}$ below $T_c$ excludes isotropic $s$ wave superconductivity in all three compounds.

PACS numbers: 74.72.Bk 74.25.Nf 75.40.Cx 76.60.Es 76.60.Gv
I. INTRODUCTION

Recently, we reported in detail nuclear magnetic resonance (NMR) and nuclear quadrupole resonance (NQR) measurements in the Y$_2$Ba$_4$Cu$_7$O$_{15}$ compound, which can be considered as a natural multilattice consisting of alternating YBa$_2$Cu$_4$O$_8$ (1–2–4 for short) and YBa$_2$Cu$_3$O$_7$ (1–2–3) blocks. The CuO$_2$ planes of adjacent 1–2–3 and 1–2–4 blocks form double planes whose individual planes are inequivalent and distinguishable by NMR/NQR. By comparing NQR frequency, spin–lattice relaxation time, $T_1$, and magnetic shift, $K$, of the distinct plane Cu sites we found three major results: (i) the individual planes of a double plane have different doping levels; (ii) the individual planes of a double plane are strongly coupled with an estimated coupling strength of at least 30 meV; (iii) the electron spin fluctuation spectrum in both individual planes exhibits a spin pseudo–gap with a common value.

To further study the interplane coupling and its consequences on inplane spin dynamics we have performed measurements of an additional NMR/NQR quantity, the Gaussian contribution of alternating YBa$_2$Cu$_4$O$_8$ and YBa$_2$Cu$_3$O$_6$.982.

The paper is organized as follows. The next section contains some necessary theoretical background. Experimental procedures, including the characterization of the sample, are given in Sec. II. In Sec. III we present our data, followed by a discussion of results in Sec. IV and a summary in Sec. V.

II. THEORY

Pennington et al. were the first to realize that the spin–spin relaxation rate of plane Cu in YBa$_2$Cu$_3$O$_7$ is much larger as expected from conventional dipolar coupling. They showed that the predominant part of the rate is due to an enhanced Cu nuclear–spin coupling induced through an indirect coupling via electron spins.

If the quantization axis of the Cu nuclear spin is parallel to the crystallographic $c$ axis, as in the case of plane copper in pure NQR, then $T_{2G,ind}$ can be expressed in terms of $\chi(q)$ as

$$\left[ \frac{1}{T_{2G,ind}} \right]^2 = \frac{P(\gamma_n \hbar)^4}{m^2 c^4} \left[ \sum_q |A(q)_{cc}|^4 \chi(q)^2 - \frac{1}{N} \sum_q |A(q)_{cc}|^2 \chi(q)^2 \right]. $$

Here, $P$ and $\gamma_n$ are the abundance and the gyromagnetic ratio of the Cu isotope being studied, $m$ is a constant that depends on the resonance method used ($m = 8$ for NMR and $4$ for NQR), $N$ is the number of Cu atoms per unit area, $c$ denotes the direction of quantization, i.e. the direction of the main component of the electric field gradient tensor in case of NQR. $A(q)$ is the Fourier transform of the hyperfine coupling tensor $A(r_i)$ consisting of the on–site $A_{cc}$ ($r_j = 0$) and isotropic transferred $B(r_j \neq 0$) terms. For the Cu nuclei under consideration and adopting the Mila–Rice Hamiltonian $A(q)_{cc}$ is given by

$$A(q)_{cc} = A_{cc} + 2B \cos(q_y a) + \cos(q_y a).$$

Since in all Y-Ba-Cu-O compounds the spin part of the plane Cu magnetic shift in $c$ direction is zero, $A_{cc} = -4B$. Consequently, $A(q)$ peaks at the corners of the first Brillouin zone at $Q_{AF} = (\pm \pi, \pm \pi, 0)$, and $T_{2G,ind}$ therefore involves predominantly $q$ summation of $\chi(q)$ close to $Q_{AF}$.

Using a phenomenological expression for $\chi(q)$, Thelen and Pines showed that, in the long correlation length limit $\xi \gg a$,

$$\frac{1}{T_{2G,ind}} \propto \frac{\chi'(Q_{AF})}{\xi} \propto \xi \sqrt{\beta},$$

where $a$ is the lattice constant and $\beta$ a parameter measuring the relative strength of the antiferromagnetic (AF) fluctuations with respect to the zone–center fluctuations.

Finally, we recall the “spin–lattice relaxation rate per temperature unit”, $(T_1 T)^{-1}$, which is given by the weighted $q$ average of the $\lim_{\omega \to 0} \chi'(q, \omega)$. For $\xi \gg a$, one obtains

$$\frac{1}{T_1 T} \propto \frac{\chi'(Q_{AF})}{\Gamma_{AF}} \sqrt{\beta},$$

where $\Gamma_{AF}$ is a characteristic AF spin fluctuation energy scale.

III. EXPERIMENT

The Y$_2$Ba$_4$Cu$_7$O$_{15}$ compound synthesized with Ba metal (not carbonate) by “method II” of Ref. was measured $T_{2G}$ by the NQR spin echo method in zero magnetic field using standard pulsed spectrometers. The signals were obtained by a phase alternating add–subtract spin–echo technique similar to that one used in Ref. To
have an optimal filling factor and thus optimal signal-to-noise ratio, we used unoriented powder, for which \( T_{2G} \) is about 7% larger than for uniaxially aligned powder. To be able to make unbiased comparison of \( Y_2Ba_2Cu_3O_7 \) \( T_{2G} \) results with those from its parent compounds, we in addition measured \( T_{2G} \) on unoriented \( YBa_2Cu_3O_7 \) and \( YBa_2Cu_3O_6.982 \) powders under the same experimental conditions.

In order to measure \( T_{2G} \) properly it is necessary to uniformly flip all nuclear spins involved in the experiment. This demands a flipping pulse \( (\pi \text{ pulse}) \) that is short compared to the inverse of the linewidth. In going from normal to superconducting state the pulse length has to be readjusted due to a changed penetration depth and an inevitable increase of the inhomogeneity of the flipping high-frequency field in the superconducting powder grains. Failing to do so results in an abrupt prolongation of \( T_{2G} \) just below \( T_c \). In general, an incomplete flipping of the spins due to whatsoever experimental shortcomings, results in a prolonged \( T_{2G} \), therefore all the presented \( T_{2G}^{-1} \) data are somewhat smaller than the proper value obtained under "ideal experimental conditions". We estimate that for the superconducting state our \( T_{2G}^{-1} \) values could be up to 20% too low because of the use of unoriented powder and the inhomogeneous high-frequency field.

The full linewidth at half height (FWHH) of the plane \( ^{63}\text{Cu} \) NQR at 100 K is 350 kHz in the 1–2–3 and 220 kHz in the 1–2–4 block of \( Y_2Ba_4Cu_7O_{15} \) (Ref. 12, and 180 kHz in \( YBa_2Cu_3O_7 \) (Ref. 11) and 200 kHz in \( YBa_2Cu_3O_6.982 \) (Ref. 12). At temperatures above \( T_c \), the length of the applied \( \pi \) pulse was 1.4 \( \mu s \), below \( T_c \) this length increased to 2.4 \( \mu s \).

IV. RESULTS AND ANALYSIS

The spin–echo amplitude \( E \), recorded as a function of time \( \tau \) between the first and the second (flipping) pulse could be fitted to the expression

\[
E(2\tau) = E_0 \exp \left[ -\frac{2\tau}{T_{2R}} - \frac{1}{2} \left( \frac{2\tau}{T_{2G}} \right)^2 \right], \tag{4.1}
\]

where the Lorentzian–Redfield term \( T_{2R}^{-1} \) stands for the decay rate due to the spin–lattice relaxation process. \( T_{2R}^{-1} \) was determined from the expression

\[
T_{2R}^{-1} = (2 + r)/3T_{1G}^{-1}, \tag{4.2}
\]

using NQR \( T_1 \) from Ref. 11. For the anisotropy of the relaxation rate, \( r \), we took the values 3.7 (Ref. 11) for \( YBa_2Cu_3O_6.982 \) and 3.3 (Ref. 12) for the 1–2–4 block and \( YBa_2Cu_3O_7 \), assuming that \( r \) is the same in the parent compound and in the corresponding block of \( YBa_2Cu_3O_7 \). With \( T_{2G} \) and \( T_{2R} \) as free fitting parameters we found that \( T_{2G} \) thus obtained agrees with that calculated from \( T_1 \). The disadvantage of the latter fitting procedure is a larger scatter of the results.

Fig. 2 presents our results for \( T_{2G}^{-1} \) of \( Y_2Ba_4Cu_7O_{15} \), \( YBa_2Cu_3O_6.982 \) and \( YBa_2Cu_4O_8 \) corrected now for “oriented powder” by multiplying the raw values by \( 1.07 \). For comparison, recent data for \( YBa_2Cu_3O_6.982 \) (multiplied by \( \sqrt{2} \) for comparison with NQR values), \( YBa_2Cu_3O_6.982 \) and \( YBa_2Cu_4O_8 \) are given. While there is rather good agreement for the \( YBa_2Cu_4O_8 \) structures, there is a discrepancy for the \( YBa_2Cu_3O_6 \) samples either for experimental reasons or because of a dependence of \( T_{2G} \) on doping (see below).

The obtained \( T_{2G}^{-1} \) encompass the temperature independent contribution, \( T_{2G,\text{ind}}^{-1} \), arising from the direct nuclear dipole–dipole interaction and the temperature dependent contribution, \( T_{2G,\text{dip}}^{-1} \), caused by the indirect nuclear spin–spin coupling mediated through the AF correlated electron spins. In a Gaussian approximation, neglecting the interference terms, both contributions add

\[
T_{2G}^{-1} \approx T_{2G,\text{dip}}^{-1} + T_{2G,\text{ind}}^{-1}. \tag{4.3}
\]

Anticipating a common temperature dependence of \( T_{2G,\text{ind}}^{-1} \) in both planes, we plotted \( (T_{2G}^{-1})_{123} \) vs. \( (T_{2G}^{-1})_{124} \) using the temperature as an implicit parameter (Fig. 2, insert). Within the experimental scatter a linear relationship

\[
(T_{2G}^{-1})_{124} = A(T_{2G}^{-1})_{123} + B \tag{4.4}
\]

with \( A = 2.01(6) \) and \( B = -17.8(8) \) ms\(^{-1}\) is indeed observed. If \( T_{2G,\text{dip}}^{-1} \) is equal in both planes, as one may expect on grounds of structural similarity, the relationship (4.4) delivers \( T_{2G,\text{dip}}^{-1} = 4.1(1.0) \) ms\(^{-1}\), which is close to the 4.5 ms\(^{-1}\) we estimate for the \( YBa_2Cu_4O_7 \) crystal structure by using a modified Abragam–Kambe expression for the NQR dipolar second moment. The \( YBa_2Cu_4O_7 \) \( T_{2G,\text{dip}}^{-1} \) value is somewhat smaller than the estimated 5.8 and 5.9 ms\(^{-1}\) obtained for \( YBa_2Cu_3O_6.982 \) and \( YBa_2Cu_3O_7 \), respectively. This difference arises from the fact that in \( YBa_2Cu_4O_7 \) the \( Cu \) NQR frequencies in the individual planes of the double plane differ, therefore the “neighbor” plane having only nonresonant \( Cu \) nuclear spins contributes much less to \( T_{2G,\text{dip}}^{-1} \) as in case of \( YBa_2Cu_3O_7 \) and \( YBa_2Cu_4O_8 \), where all planes are equivalent.

Finally, Eq. (4.2) yields \( T_{2G,\text{ind}}^{-1} \), the results are plotted in Fig. 2. Notice that \((1/T_{2G,\text{ind}}^{-1})_{124} > (1/T_{2G,\text{ind}}^{-1})_{123}\), meaning that the planes of the 1–2–4 block are less doped than those of the 1–2–3 block, in agreement with our earlier conclusions. Further, \( T_{2G,\text{ind}}^{-1} \) in both blocks of \( YBa_2Cu_4O_7 \) follows the same temperature dependence above and below \( T_c \), as seen from the constant ratio \( r_{2R} = T_{2G,\text{ind}}^{-1}/T_{2G,\text{dip}}^{-1} \) (Fig. 2, insert).

Around 50 K, we observe in \( YBa_2Cu_4O_7 \) at both plane \( Cu \) sites an unexpected growth of \( T_{2R}^{-1} \), peaking at 52 K. Similar peaks were detected at 87 K in \( YBa_2Cu_3O_6.982 \) and \( YBa_2Cu_3O_7 \). Since no such peaks appear in \( YBa_2Cu_4O_8 \) which stands out in the \( Y–Ba–Cu–O \) family as a stoichiometric, thermally very stable
compound, we suspect that the diffusion of loosely bound oxygen in the single chains, known as the weak structural elements present in YBa$_2$Cu$_3$O$_7$ and Y$_2$Ba$_4$Cu$_7$O$_{15}$ but not in YBa$_2$Cu$_3$O$_6$, could be the source of this $T_{2R}$ anomaly. A more detailed study to clarify this aspect is necessary. However, we believe that this anomaly is extrinsic rather than a genuine effect of the inplane electron spin dynamics.

V. DISCUSSION

The experimental results on $T_{2G}$ allow us to discuss five features which characterize Y$_2$Ba$_4$Cu$_7$O$_{15}$ at the microscopic level: the interplane coupling, the temperature dependence of the electronic susceptibility at the AF wave vector, the spin gap and its relation to the interplane coupling and, finally, the symmetry of the pair wave function of the superconducting state.

A. Further evidence for interplane coupling

The presence of interplane coupling, at least for the normal conducting phase of Y$_2$Ba$_4$Cu$_7$O$_{15}$, were deduced from the fact that the spin–lattice relaxation rate for the two inequivalent plane Cu sites exhibited the same temperature dependence; the same was found for the Knight shift at these sites. The temperature dependence itself shows a behavior typical for an underdoped high-T$_c$ compound that forms a spin gap (see below). Since both the relaxation rate and the Knight shift are related to the dynamic susceptibility, the common temperature dependence reveals the same dynamics in these planes which must arise from a coupling between the planes.

As shown by the insert of Fig. 2, also the $T_{2G}^{-1}$ data of the two inequivalent Cu sites reveal a common temperature dependence and thus provide a third piece of evidence for the interplane coupling.

B. Temperature dependence of $\chi(Q_{AF})$ above $T_c$

In principle, using experimental $T_{2G}$ and $T_1$ data, the electronic susceptibility at the AF wave vector, in the long correlation length limit, can be derived from Eqs. (2.3) and (2.4), respectively. This requires, however, some knowledge about the temperature dependence of the parameters $\xi$ and $\beta$ which is not known a priori. Nevertheless, similar to the treatment of YBa$_2$Cu$_3$O$_{6.9}$ in Ref. 11 we can discuss two limiting cases. (i) If the correlation length, $\xi$, is independent of temperature, $\chi'(T)$ is determined by the temperature dependence of $T_{2G,ind}$ according to Eq. (2.3), hence $\chi'(Q_{AF}) \propto 1/T_{2G,ind}$. (ii) If on the other hand, $\xi$ depends strongly on temperature and $\beta$ does not, we have $1/T_{2G,ind} \propto \xi$ and $\chi'(Q_{AF}) \propto \xi^2$, hence $\chi'(Q_{AF}) \propto T_{2G,ind}^{-2}$.

In Fig. 2 we have plotted the temperature dependence of both $T_{2G,ind}$ and $T_{2G,ind}^2$ corresponding to cases (i) and (ii), respectively, together with our data for YBa$_2$Cu$_3$O$_{6.982}$ and YBa$_2$Cu$_3$O$_6$. For case (i), all four data sets can be fitted by a Curie-Weiss law (dotted curves) which implies that $1/\chi'(Q_{AF}) \propto 1/(T + \Theta)$ where $\Theta$ is the Weiss temperature. For case (ii), the inverse susceptibility of the 1–2–3 and the 1–2–4 blocks of Y$_2$Ba$_4$Cu$_7$O$_{15}$ follows a Curie law, i.e., $1/\chi'(Q_{AF}) \propto 1/T$, while the YBa$_2$Cu$_3$O$_7$ and YBa$_2$Cu$_4$O$_8$ data show a different behavior.

We believe that case (i), i.e., a temperature independent or at least only weakly dependent correlation length, is the correct interpretation of the data since it applies to all four data sets with a fit quality that is better than in case (ii). Furthermore, this result is in accord with theoretical models and supports results of inelastic neutron scattering in YBa$_2$Cu$_4$O$_8$.

Accepting this conclusion, we note that the negative value of the Weiss temperatures imply that the compounds do not order anti-ferromagnetically at low temperatures which is a well-known fact. For the two blocks of Y$_2$Ba$_4$Cu$_7$O$_{15}$, the Weiss temperatures are the same (about $-200$ K) while $\Theta$ is $-100$ K and $-300$ K in the YBa$_2$Cu$_3$O$_6$ and YBa$_2$Cu$_4$O$_8$ structures, respectively. Thus, the absolute value of the Weiss temperature in the Y-Ba-Cu-O compounds increases with rising doping level and distinguishes Y$_2$Ba$_4$Cu$_7$O$_{15}$ as a (nearly) optimized compound. A similar behavior, according to $T_1$ measurements had been found for La-Sr-Cu-O compounds where the Sr content determines the doping level.

C. Spin gap behavior and interplane coupling

Next, we will discuss the energy scale parameter of the AF fluctuations, $\Gamma_{AF}$, and the evidence for the presence of a spin gap. NMR investigations of underdoped YBa$_2$Cu$_3$O$_x$ compounds of YBa$_2$Cu$_4$O$_8$ (Ref. 23) and of Y$_2$Ba$_4$Cu$_7$O$_{15}$ (Ref. 1) have shown that the planar Cu spin–lattice relaxation and Knight shift data can be interpreted in terms of a spin gap, in accord with neutron scattering data. The occurrence of a spin gap means that spectral weight in the electron spin fluctuations is transferred from lower to higher energy. The presence of the spin gap manifests itself in a maximum of $1/T_1 T$ at a temperature $T^*$ well above $T_c$, with $T^*$ = 130 and 150 K for Y$_2$Ba$_4$Cu$_7$O$_{15}$ (Ref. 1) and YBa$_2$Cu$_4$O$_8$ (Ref. 25), respectively. It should be stressed again that the $T_{2G}^{-1}$ data do not show such a peak.

Combining the Eq. (2.3) and (2.4) yields the quantity $T_1 T/T_{2G,ind}$ which is proportional to $\Gamma_{AF}$. The plot of Fig. 2 reveals that for both blocks of Y$_2$Ba$_4$Cu$_7$O$_{15}$ and for YBa$_2$Cu$_4$O$_8$, $\Gamma_{AF}$ increases with falling temperature whereas for YBa$_2$Cu$_3$O$_{6.982}$ it remains constant. The
latter result is in agreement with earlier measurements by the Slichter group \(^1\), our YBa\(_2\)Cu\(_4\)O\(_8\) data are similar to those of Itoh et al. \(^1\).

In the framework of the random phase approximation (RPA) formalism \(^1\), one does not expect such an increase of \(\Gamma_{AF}\), instead the spin fluctuations are supposed to be slowed down by growing AF correlation on decreasing temperature. However, if the spin excitation spectrum is changed by the opening of a spin gap, the increase of \(\Gamma_{AF}\) is conceivable.

The origin of the spin gap is still under debate. For instance, one may ask whether the gap is an intrinsic property of the single CuO\(_2\) plane or a consequence of interplane effects. One approach to answer this question \(^2\) considers the spin gap an intrinsic property of a single underdoped CuO\(_2\) plane that shows a quasi twodimensional quantum Heisenberg antiferromagnet behavior. The spin gap itself is related to the suppression of spectral weight for frequencies smaller than \(v_s/\xi\) (where \(v_s\) is the spin wave velocity) in the quantum disordered regime. Another approach \(^3\) assumes that the gap originates from an effective interplane coupling between adjacent CuO\(_2\) planes.

Pursuing the first path, Sokol and Pines (SP) \(^4\) propose an unified magnetic phase diagram of the cuprate superconductors, which, dependent on doping level and temperature, displays various regimes that, among others, are characterized by certain temperature independent ratios of plane Cu \(T_{J1}T\) and \(T_{2G,ind}\) values. In the quantum critical (QC) regime (applicable to spin gap compounds), the ratio \(T_{J1}T/T_{2G,ind}\) is constant while in the overdamped (OD) regime \(T_{J1}T/T_{2G,ind}\) is constant.

We check these predictions with the help of Fig. \(^1\) and its insert. Obviously, the overdoped compound YBa\(_2\)Cu\(_3\)O\(_7\) is in the OD regime from \(T_c\) up to 300 K (as noted already by SP, although only for the 150–300 K range). On the other hand, the underdoped YBa\(_2\)Cu\(_4\)O\(_8\) structure is in the QC regime for temperatures above \(T_{J1}\).

While these results are in accord with the SP model, the Y\(_2\)Ba\(_2\)Cu\(_7\)O\(_{15}\) structure does not fit into this scheme. The mere fact that the differently doped planes of the 1–2–3 and 1–2–4 blocks have the same cross-over temperature \(T_{2G}\) already contradicts the proposed phase diagram. In particular, above \(T_{2G}\), the two blocks do not show the \(T_{J1}T/T_{2G,ind} = \text{constant}\) behavior expected for the QC regime the material should belong to because of the spin gap. Instead, we note that \(T_{J1}T/T_{2G,ind} = \text{constant}\) which is the signature of the OD regime.

On the other hand, the second approach, by taking the interplane coupling as the origin of the spin gap, quite naturally accounts for the observed Y\(_2\)Ba\(_2\)Cu\(_7\)O\(_{15}\) behavior. The interplane coupling not only dictates a common temperature dependence of the dynamic susceptibility in adjacent planes, it also leads to a \(\text{common}\) value of the spin gap in the fluctuation spectrum. If we make the reasonable assumption that the effective interplane coupling depends on the sum of charge carrier effects in both planes, one understands why the cross-over temperature of Y\(_2\)Ba\(_4\)Cu\(_7\)O\(_{15}\), \(T^* = 130\) K, lies somewhere between the corresponding values for YBa\(_2\)Cu\(_3\)O\(_7\) (\(T^* \leq T_c\)) and YBa\(_2\)Cu\(_4\)O\(_8\) (\(T^* = 150\) K).

**D. Symmetry of superconducting state**

We now discuss the relevance of our results with respect to the symmetry of the pair wave function of the superconducting state. We therefore have plotted in Fig. \(^5\) \(T_{2G,ind}(T)/T_{2G,ind}(T_c)\) vs \(T/T_c\) for all plane Cu sites in Y\(_2\)Ba\(_4\)Cu\(_7\)O\(_{15}\), YBa\(_2\)Cu\(_4\)O\(_8\) and YBa\(_2\)Cu\(_3\)O\(_{6.982}\). Within error limits the various data points gather on an “universal” curve which decreases monotonously towards 0.80 in the \(T \to 0\) limit. According to the discussion of Sec. \(^3\), the “true” limiting value of \(T_{2G,ind}(T)/T_{2G,ind}(T_c)\) could be about 20% higher.

The dashed and dotted lines in Fig. \(^6\) are theoretical curves calculated by Bulut and Scalapino \(^7\) for YBa\(_2\)Cu\(_3\)O\(_7\) in case of s and d wave symmetry. Although the corresponding calculations for Y\(_2\)Ba\(_4\)Cu\(_7\)O\(_{15}\) and YBa\(_2\)Cu\(_4\)O\(_8\) have not yet been performed it is reasonable to assume that these “normalized” curves will be not much different from the YBa\(_2\)Cu\(_3\)O\(_7\) curves since the experimental data for all compounds follow an universal curve.

The experimental points are much closer to the \(d\) than the \(s\) wave curve, in particular if a possible 20% correction is taken into account. Hence, the \(T_{2G,ind}\) data seem to favor \(d\) wave symmetry. For YBa\(_2\)Cu\(_4\)O\(_8\), this result is in accord with our previous conclusion drawn from Cu spin–lattice relaxation and Knight shift measurements \(^8\). However, as shown by Sudbo et al. \(^9\) models of \(d\) wave and strongly anisotropic \(s\) wave superconductivity deliver very similar results. Thus, we conclude that the \(T_{2G}\) data exclude \(isotropic\) \(s\) wave symmetry.

**VI. SUMMARY**

We have presented plane Cu nuclear spin–spin relaxation rates, \(T_2^{-1}\), of Y\(_2\)Ba\(_4\)Cu\(_7\)O\(_{15}\), YBa\(_2\)Cu\(_3\)O\(_8\) and YBa\(_2\)Cu\(_3\)O\(_{6.982}\), which deliver information on the static electron spin susceptibility, \(\chi'(q)\), at non–zero wave vector, \(q\). Our main conclusions were drawn from a discussion of the indirect component, \(T_{2G,ind}\), of the Gaussian contribution to \(T_2\).

For Y\(_2\)Ba\(_4\)Cu\(_7\)O\(_{15}\), the magnitude of the individual \(T_{2G,ind}\) values confirms our previous conclusion \(^1\) that the planes of the 1–2–4 block are less doped than the 1–2–3 block planes. The temperature dependence of \(T_{2G,ind}\) provides further evidence for a strong interplane coupling between adjacent CuO\(_2\) planes belonging to different blocks.

For all three compounds studied, the data suggest a nearly temperature independent correlation length of the
anti-ferromagnetic (AF) fluctuations and a Curie–Weiss law for the susceptibility at the AF wave vector with negative Weiss temperatures whose absolute values increase with doping level.

The data support our previous conclusion about the existence of the spin pseudo–gap in $Y_2Ba_4Cu_7O_{15}$ and $YBa_2Cu_4O_8$. The observation of the same gap value for both blocks in $Y_2Ba_4Cu_7O_{15}$ points to the importance of the interplane coupling in the gap formation. We show that the $Y_2Ba_4Cu_7O_{15}$ results are incompatible with the Sokol—Pines phase diagram based on single CuO$_2$ plane theory while the $YBa_2Cu_4O_8$ and $YBa_2Cu_3O_6.982$ data are in accord with that theory.

The temperature dependence of $T_{2G,ind}$ below $T_c$ excludes isotropic s wave superconductivity in $Y_2Ba_4Cu_7O_{15}$, $YBa_2Cu_4O_8$ and $YBa_2Cu_3O_6.982$. Together with our spin–lattice relaxation and Knight shift measurements in $YBa_2Cu_4O_8$ (Ref. 25) and corresponding investigations of $YBa_2Cu_3O_7$ (Ref. 33), one may conclude that all Y–Ba–Cu–O compounds are not isotropic s wave superconductors. At present it seems impossible to distinguish between possible d and more exotic s wave superconductivity in these materials.

ACKNOWLEDGMENTS

We thank the group of Prof. E. Kaldis (ETH–Zurich) for preparing the $Y_2Ba_4Cu_7O_{15}$ material. Financial support by the Swiss National Science Foundation is gratefully acknowledged.

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* Also at the Institute of Chemical Physics and Biophysics, EE–0001 Tallinn, Estonia.
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FIG. 1. Temperature dependence of NQR \( T_G^{-1} \) at the plane copper sites in the 1–2–3 (○) and 1–2–4 block (●) of Y\(_2\)Ba\(_3\)Cu\(_7\)O\(_{15}\), in YBa\(_2\)Cu\(_3\)O\(_{6.98}\) (□), and YBa\(_2\)Cu\(_4\)O\(_8\) (■). For comparison: data for YBa\(_2\)Cu\(_3\)O\(_{6.9}\) (△, joined with dotted line, Ref. 16), YBa\(_2\)Cu\(_3\)O\(_{6.98}\) (dash–dotted line, Ref. 17) and YBa\(_2\)Cu\(_4\)O\(_8\) (dashed line, Ref. 18). Insert: \( T_G^{-2} \) of 1–2–4 block vs that of 1–2–3 block with temperature as an implicit parameter.

FIG. 2. Temperature dependences of NQR \( T_G^{-1} \)\(_{ind} \) at the plane copper sites. The symbols are the same as in Fig. 1. Insert: ratio of \( T_G^{-1} \)\(_{ind} \) of individual planes in Y\(_2\)Ba\(_3\)Cu\(_7\)O\(_{15}\).

FIG. 3. Temperature dependences of (a) \( T_G^{-1} \)\(_{ind} \) and (b) \( T_G^{2} \)\(_{ind} \). The symbols are the same as in Fig. 1. Dotted lines are fits to the (a) Curie–Weiss or (b) Curie law.

FIG. 4. Temperature dependences of \( T_1/T_G^{2} \)\(_{ind} \) (\( \propto \gamma_{AF} \)) and \( T_1/T_G^{-1} \)\(_{ind} \) (insert). The symbols are the same as in Fig. 1. The lines are guides to the eye, the arrows mark \( T_c \) and \( T^* \) of Y\(_2\)Ba\(_3\)Cu\(_7\)O\(_{15}\).

FIG. 5. Temperature dependences of \( T_G^{-1} \)\(_{ind} \) at the plane copper sites in the superconducting state. The symbols are the same as in Fig. 1 the solid line is a guide to the eye. The dashed and dotted lines are calculated values (Ref. 31) for a \( s \) and a \( d \) wave superconductor, respectively.