Inter-plane Coupling and Spin Gap – an NMR/NQR Look on Typical Properties of High-Temperature Superconductors *

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Z. Naturforsch. **51a**, 786–792 (1996); received November 20, 1995

The paper discusses some NQR/NMR studies performed on Y–Ba–Cu–O superconductors at the University of Zürich. In particular, we review studies performed in $Y_2Ba_4Cu_7O_{15}$ by measuring various planar Cu NQR/NMR parameters: the spin-lattice relaxation time, the Knight shift and the indirect component of the Gaussian contribution to the spin-spin relaxation time. The temperature dependence of these parameters reveals a coupling between adjacent planes of a double plane. The existence of the inter-plane coupling has independently been confirmed by performing NQR Spin-Echo Double Resonance (SEDOR) experiments. The appearance of a spin gap seems to be the consequence of inter-plane coupling.

Key words: NQR, NMR, High-Temperature Superconductors, Inter-plane Coupling, Spin gap.

1. Introduction

Up to now, an impressive number of nuclear magnetic resonance (NMR) and nuclear quadrupole resonance (NQR) experiments on planar copper and oxygen nuclei in cuprate high-temperature superconductors has been performed to elucidate the electronic spin dynamics of the CuO_2 planes where superconductivity takes place. In most of these works, however, the aspect of the inter-plane coupling and its influence on the spin dynamics has not been addressed, even though neutron scattering experiments in Y–Ba–Cu–O structures [1] revealed strong antiferromagnetic (AF) correlations between spins in different planes of the double plane. However, at elevated temperatures magnetic neutron scattering becomes extremely weak in the normal conducting state of $YBa_2Cu_3O_x$, in particular when x is close to 7. Therefore, an alternative study of the inter-plane coupling in this temperature region by NMR/NQR could be of great help, especially with regard to the possible connection between inter-plane coupling and the spin pseudo-gap.

Such NMR/NQR studies became feasible with the synthesis of $Y_2Ba_4Cu_7O_{15}$ ($T_c = 93$ K), whose structure consists of a sequence of alternating $YBa_2Cu_3O_7$ (1–2–3 for short) and $YBa_2Cu_4O_8$ (1–2–4) blocks containing single and double CuO chains, respectively. As in $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$, the CuO_2 planes in $Y_2Ba_4Cu_7O_{15}$ form double planes separated by Y ions. However, due to the alternation of 1–2–3 and 1–2–4 blocks the individual planes of a CuO_2 double plane in $Y_2Ba_4Cu_7O_{15}$ are *inequivalent* and hence can separately be monitored by NMR/NQR methods allowing one to investigate the coupling between these planes. We will report on such studies performed in our laboratory.

* Presented at the XIIIth International Symposium on Nuclear Quadrupole Interactions, Providence, Rhode Island, USA, July 23–28, 1995.

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2. Probing the Dynamical Electron Susceptibility

The electronic spin fluctuations in the Y–Ba–Cu–O superconductors play an essential role. The low-frequency spectrum of these fluctuations can be studied by probing the wave-vector (q) and frequency (ω) dependent spin susceptibility, $\chi(q, \omega)$. Relevant information can be gained from the temperature dependence of three parameters: the magnetic shift tensor, the nuclear spin-lattice relaxation time, T_1, and the Gaussian contribution, T_{2G}, to the spin-spin relaxation time [2–4]. We briefly summarize how these parameters are related to $\chi(q, \omega)$.

The static part of the local magnetic hyperfine field gives rise to an NMR line shift expressed by the *magnetic shift tensor* **K**, whose components, in an x, y, z reference frame, can be decomposed in a spin and an

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orbital part:

$$K_{\alpha\alpha}(T) = K_{\alpha\alpha}^{\text{spin}}(T) + K_{\alpha\alpha}^{\text{orb}}. \quad (1)$$

The x, y, z reference frame quite often coincides with the principal axes system of the electric field gradient (EFG) tensor, $\mathbf{V}$, at the nuclear site. The spin part of the magnetic shift is usually called the Knight shift. In the high-T_c compounds, $\mathbf{K}^{\text{orb}}$ is predominantly temperature independent, whereas the temperature dependent $\mathbf{K}^{\text{spin}}$ is expected to vanish in the superconducting state due to singlet spin pairing.

Each part of the $\mathbf{K}$ tensor can be expressed by the respective hyperfine interaction tensor and the static electronic susceptibility as

$$K_{\alpha\alpha}^{\text{spin}} = \frac{1}{g\,\mu_B} \sum_j (A_j)_{\alpha\alpha} (\chi_j)_{\alpha\alpha}, \quad (2)$$

$$K_{\alpha\alpha}^{\text{orb}} = \frac{1}{\mu_B} O_{\alpha\alpha} \chi_{\alpha\alpha}^{\text{orb}}. \quad (3)$$

The fluctuating part of the local hyperfine field is the source of the nuclear spin-lattice relaxation. In case of high-T_c compounds, the main contribution to the copper spin-lattice relaxation stems from the electron spin fluctuations. After Moriya [5], this contribution is related to the imaginary part of $\chi(q,\omega)$ in the following way:

$$\left(\frac{1}{T_1 T}\right)_\alpha = \frac{\gamma_n^2 k_B}{2\mu_B^2} \sum_{q,\alpha' \neq \alpha} |A(q)_{\alpha'\alpha'}|^2 \frac{\chi''_{\alpha'\alpha'}(q,\omega_0)}{\omega_0}, \quad (4)$$

$$A(q)_{\alpha\alpha} = \sum_j A_{j,\alpha\alpha} \exp(i\,\mathbf{q}\cdot\mathbf{r}_j).$$

Here, ω_0 is the nuclear resonance frequency. α denotes the direction of quantization, i.e. the direction of V_{zz} in NQR and of $\mathbf{B}_0$ (the external magnetic field) in NMR experiments, and α' is the direction perpendicular to α. $\mathbf{A}_j$ is the on-site ($\mathbf{r}_j = 0$) and the transferred ($\mathbf{r}_j \neq 0$) hyperfine coupling tensor for the nuclei under consideration. Thus, the "relaxation rate per temperature unit" provides information about the q averaged imaginary part of $\chi(q,\omega_0)$.

Pennington et al. [6] were the first to realize that the spin-spin relaxation rate of plane Cu in $YBa_2Cu_3O_7$ is much larger than expected from conventional nuclear dipolar coupling. They showed that the predominant part of the rate is due to an enhanced Cu nuclear-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 $T_{2G,\text{ind}}^{-1}$ can be expressed [7, 8] in terms of $\chi'(q)$ as

$$\left[\frac{1}{T_{2G,\text{ind}}}\right]^2 = \frac{P}{m\hbar^2}\left[\frac{1}{N}\sum_q |A(q)_{cc}|^4 \chi'(q)^2 - \left[\frac{1}{N}\sum_q |A(q)_{cc}|^2 \chi'(q)\right]^2\right]. \quad (5)$$

Here, P is the abundance 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, and c denotes the direction of quantization, i.e. the direction of the main component of the EFG tensor in case of NQR. $\mathbf{A}(q)$ is again the Fourier transform of the hyperfine coupling tensor $\mathbf{A}(r_i)$ consisting of the on-site A_{cc} ($\mathbf{r}_j = 0$) and isotropic transferred B ($\mathbf{r}_j \neq 0$) terms. For the Cu nuclei under consideration, and adopting the Mila-Rice Hamiltonian [9], $A(q)_{cc}$ is given by

$$A(q)_{cc} = A_{cc} + 2B[\cos(q_x a) + \cos(q_y a)]. \quad (6)$$

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, $\mathbf{A}(q)$ peaks at the corners of the first Brillouin zone at the AF wave-vector $Q_{AF} = (\pm\frac{\pi}{a}, \pm\frac{\pi}{a})$, and $T_{2G,\text{ind}}^{-1}$ therefore involves predominantly q summation of $\chi'(q)$ close to Q_{AF}.

3. Measurement of Knight Shift and Relaxation in $Y_2Ba_4Cu_7O_{15}$

We are now going to present the results of our NMR/NQR measurements in $Y_2Ba_4Cu_7O_{15}$.

3.1 *Knight shift*

Details of the evaluation of the Knight shift from the Cu NMR spectrum were given in [10]. For $\mathbf{B}_0 \perp \mathbf{c}$, we were able to extract, from the Cu spectrum, the temperature dependence of the total shift $K_{aa} (= K_{bb})$ for Cu(2) of the 1–2–3 block and Cu(3) of the 1–2–4 block. However, to obtain the more relevant temperature dependent spin part $\mathbf{K}^{\text{spin}}$ one has to know the constant orbital part $\mathbf{K}^{\text{orb}}$, usually obtained as the rest $\mathbf{K}$ shift at temperatures far below T_c. In case of $Y_2Ba_4Cu_7O_{15}$, the broadening and the partial overlap of the two Cu lines prevents an accurate enough determination of $\mathbf{K}^{\text{orb}}$. Therefore we had to resort on the values obtained by Barret et al. for $YBa_2Cu_3O_7$ [11] and by Zimmermann et al. for $YBa_2Cu_4O_8$ [12].

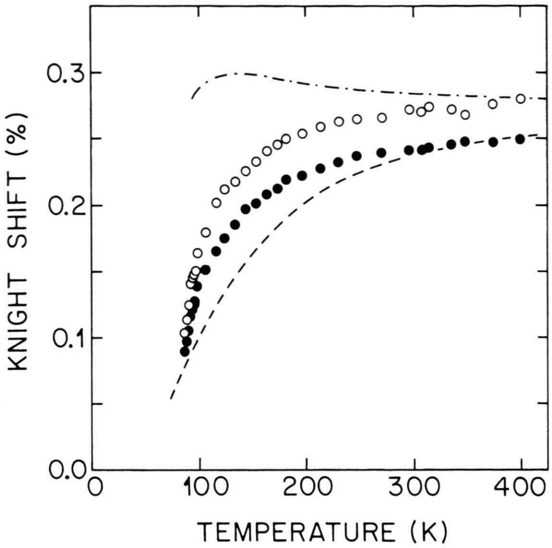

Fig. 1. Temperature dependence of the planar ^{63}Cu Knight shift at Cu(2) (open circles) and Cu(3) sites (filled circles) in $Y_2Ba_4Cu_7O_{15}$ [10] compared to the Knight shift at Cu(2) sites in $YBa_2Cu_3O_7$ (dash-dotted line, [25] and in $YBa_2Cu_4O_8$ (dashed line, [12]). The external field $\mathbf{B}_0$ lies perpendicular to the crystal **c** axis.

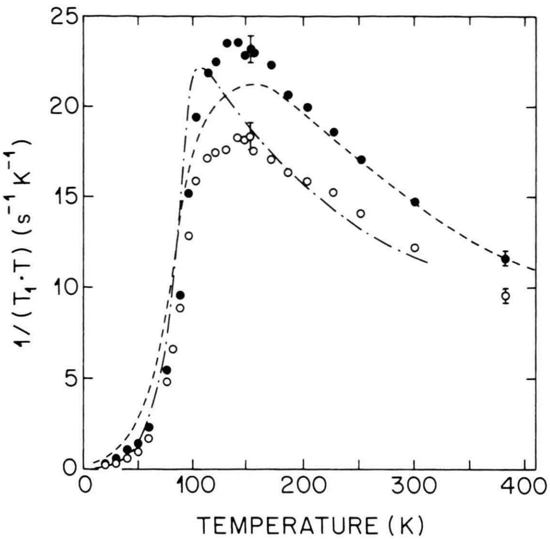

Fig. 2. Temperature dependence of ^{63}Cu $(T_1\,T)^{-1}$ measured by NQR at Cu(2) (open circles) and Cu(3) sites (filled circles) in $Y_2Ba_4Cu_7O_{15}$ [10] compared to $(T_1\,T)^{-1}$ at Cu(2) sites in $YBa_2Cu_3O_7$ (dash-dotted line, [26]) and in $YBa_2Cu_4O_8$ (dashed line, [27]).

As one may expect both values agree in error limits. Thus we took the average value of 0.285% for $K_{aa}^{\text{orb}}(=K_{bb}^{\text{orb}})$ of Cu(2) and Cu(3). Subtraction of K_{aa}^{orb} from the respective total K_{aa} then yields the temperature dependent K_{aa}^{spin} for Cu(2) and Cu(3) (see Figure 1).

3.2 Spin-lattice relaxation

The spin-lattice relaxation rate has been measured by NQR for all four copper sites of $Y_2Ba_4Cu_7O_{15}$. Figure 2 depicts the temperature dependence of $(T_1\,T)^{-1}$ of Cu(2) and Cu(3) together with related data from $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$. Characteristic for $Y_2Ba_4Cu_7O_{15}$ is that $(T_2\,T)^{-1}$ for both planar sites depends strongly on temperature in the normal conducting state showing a maximum at $T^* = 130$ K combined with a Curie-Weiss type behavior above and a strong drop below T^*.

After the discovery of the spin-gap effect [13] in $YBa_2Cu_3O_{7-\delta}$ by neutron scattering [14] several NMR groups have regarded the spin-gap effect to be responsible for the peculiar temperature variation of the relaxation rate, at least in the normal conducting state [4]. 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. Therefore, the relaxation rate is described by an *ad hoc* formula:

$$\frac{1}{T_1\,T} = \left(\frac{A_0}{T}\right)^{\alpha}\left[1-\tanh^2\left(\frac{\Delta_{\text{AF}}}{2\,T}\right)\right]. \tag{7}$$

Here, Δ_{AF} denotes the spin-gap energy at Q_{AF} since the Cu relaxation is dominated by the strong AF fluctuations around Q_{AF}. A_0 is a constant and the factor $T^{-\alpha}$ guarantees a reasonable description for the high-temperature behavior and may by attributed to the gradual decay of AF correlations at higher temperatures.

3.3 Spin-spin relaxation

The evaluation of $T_{2\text{G}}$ and hence of $T_{2\text{G,ind}}$ is more involved [15]. $T_{2\text{G}}$ follows from the decay of the spin-echo amplitude, $E(2\,\tau)$, recorded as a function of time τ between the first and the second pulse:

$$E(2\,\tau) = E_0 \exp\left[-\frac{2\,\tau}{T_{2\text{R}}} - \frac{1}{2}\left(\frac{2\,\tau}{T_{2\text{G}}}\right)^2\right], \tag{8}$$

where the Lorentzian-Redfield term T_{2R}^{-1} stands for the decay rate due to the spin-lattice relaxation process. Details how to determine T_{2R}^{-1} can be found in [15].

The obtained T_{2G}^{-1} encompasses the temperature independent contribution, $T_{2G,\mathrm{dip}}^{-1}$, arising from the direct nuclear dipole-dipole interaction and the temperature dependent contribution, $T_{2G,\mathrm{ind}}^{-1}$, 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 as [18]

$$T_{2G}^{-2} \approx T_{2G,\mathrm{dip}}^{-2} + T_{2G,\mathrm{ind}}^{-2}. \tag{9}$$

Anticipating a common temperature dependence of $T_{2G,\mathrm{ind}}^{-1}$ in both planes of $Y_2Ba_4Cu_7O_{15}$, we plotted $(T_{2G}^{-2})_{124}$ vs. $(T_{2G}^{-2})_{123}$ using the temperature as an implicit parameter. Within the experimental scatter a linear relationship $(T_{2G}^{-2})_{124} = A(T_{2G}^{-2})_{123} + B$ is indeed observed.

Figure 3 displays our results for $T_{2G,\mathrm{ind}}$ in $Y_2Ba_4Cu_7O_{15}$, $YBa_2Cu_3O_{6.982}$ and $YBa_2Cu_4O_8$. For comparison, recent data for $YBa_2Cu_3O_{6.9}$ [17], $Yba_2Cu_3O_{6.98}$ [18] and $YBa_2Cu_4O_8$ [16] are given. While there is rather good agreement for the $YBa_2Cu_4O_8$ structures, there is a discrepancy for the $YBa_2Cu_3O_x$ samples either for experimental reasons or because of a dependence of T_{2G} on doping.

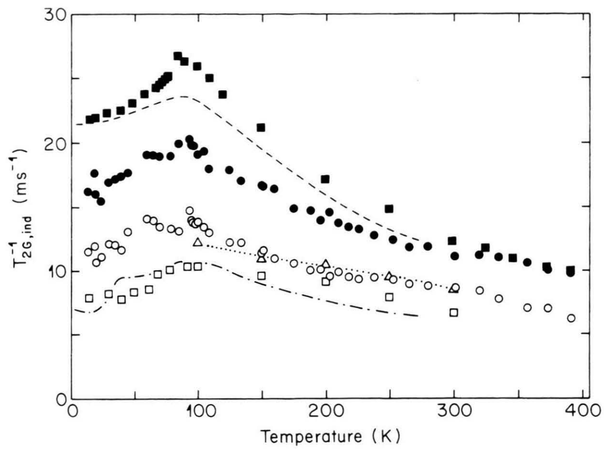

Fig. 3. Temperature dependence of NQR $T_{2G,\mathrm{ind}}^{-1}$ at the planar copper sites in the 1–2–3 (○) and 1–2–4 blocks (●) of $Y_2Ba_4Cu_7O_{15}$, in $YBa_2Cu_3O_{6.982}$ (□), and $YBa_2Cu_4O_8$ (■) [15]. For comparison, data for $YBa_2Cu_3O_{6.9}$ (△, joined with dotted line, [17]), $YBa_2Cu_3O_{6.98}$ (dash-dotted line, [18]), and $YBa_2Cu_4O_8$ (dashed line, [16]).

4. Inter-plane Coupling in $Y_2Ba_4Cu_7O_{15}$

The most important result of the previous section is summarized in Figure 4. It shows the temperature dependence of three ratios of NMR/NQR parameters of the two inequivalent planes in $Y_2Ba_4Cu_7O_{15}$: (i) Knight shifts, $r_K = K_{aa}^{\mathrm{spin}}[\mathrm{Cu(3)}]/K_{aa}^{\mathrm{spin}}[\mathrm{Cu(2)}]$; (ii) spin-lattice relaxation rates, $r_W = W[\mathrm{Cu(3)}]/W[\mathrm{Cu(2)}]$; (iii) the indirect part to the Gaussian contribution to the spin-spin relaxation rate, $r_{T2} = T_{2G,\mathrm{ind}}^{-1}[\mathrm{Cu(3)}]/T_{2G,\mathrm{ind}}^{-1}[\mathrm{Cu(2)}]$.

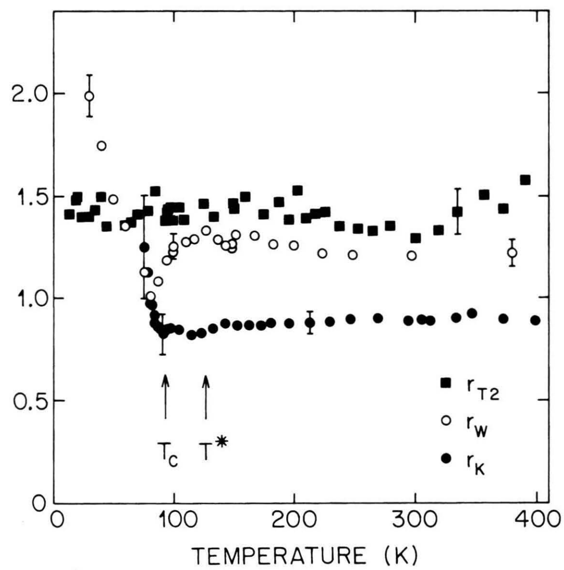

Fig. 4. Temperature dependence of the ratio of Cu(3) and Cu(2) NMR/NQR parameters in $Y_2Ba_4Cu_7O_{15}$: Knight shifts (filled circles), spin-lattice relaxation rates (empty circles) and indirect components of the Gaussian contribution to the spin-spin relaxation rate (squares).

Above 100 K, all three ratios are constant, the ratio r_{T2} is constant even above 14 K. According to (2), (4) and (5), the temperature independent ratios imply a temperature independent relationship between the respective static and dynamic electronic spin susceptibilities. The relaxation rates, strictly speaking, involve q average of $\chi(q, \omega_0)$. However, if we assume that the spin fluctuations at the corner of the Brillioun zone, $q = Q_{\mathrm{AF}}$, dominate the relaxation, the sum over q can be replaced by $\chi''(Q_{\mathrm{AF}})$ and $\chi'(Q_{\mathrm{AF}})$, respectively. The plane copper hyperfine constants appearing in the ratios are alike in Y–Ba–Cu–O compounds [19, 20] and seem to be temperature independent. In other words: both the static and the dynamic electron susceptibilities of the two single planes of each double-plane are governed by the *same* temperature dependence which means common spin dynamics in both planes, and hence these planes must be strongly coupled. However, since r_K, r_W and r_{T2} are not equal, the q dependence of the susceptibility is not the same in the two planes.

The coupling *strength* may be estimated from the K_{aa}^{spin}-vs.-temperature plot (Figure 1). Below 300 K, the K_{aa}^{spin} shifts in the 1–2–3 and 1–2–4 blocks begin to depart from those in $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$ due to the interplane coupling, thus the coupling strength is not much less than $k_B \cdot 300$ K or 30 meV.

Why is the common temperature dependence of the Knight shift and of the spin-lattice relaxation rate lost below T_c? A loss of the inter-plane coupling is unlikely. A possibility could be the formation of two superconducting gaps that differ in the individual planes because of their different charge carrier densities, n. However, up to now even the spatial symmetry of the superconducting state in Y–Ba–Cu–O, and so the form of the superconducting gap is not well established, let alone its dependence on n. Thus, the explanation of the temperature variation of r_W and r_K below T_c is still missing.

5. SEDOR experiments in $Y_2Ba_4Cu_7O_{15}$

We have performed NQR spin-echo double resonance (SEDOR) experiments in $Y_2Ba_4Cu_7O_{15}$ [21] to confirm independently and directly the existence of the inter-plane coupling and to determine the temperature dependence of the resulting inter-plane component of the static electron spin susceptibility in the normal state. To our knowledge, it is the first SEDOR experiment performed in $Y_2Ba_4Cu_7O_{15}$ and the first in high-T_c superconductors involving nuclei in different planes. Observing a SEDOR proves that the two nuclei involved are coupled by a spin-spin interaction [22].

The SEDOR experiment proceeds as follows. We perform a spin-echo experiment on nuclear spin I, e.g. the Cu(2) spin, by applying a $\pi/2-\tau-\pi$ radio-frequency pulse sequence at the NQR frequency of I and observing the echo of the I spins; τ denotes the time separation of the two pulses. In addition we apply, at time τ_F after the $\pi/2$ pulse, another π pulse at the frequency of spin S, e.g. the Cu(3) spin. This pulse flips the S spin and thus changes the local field produced by

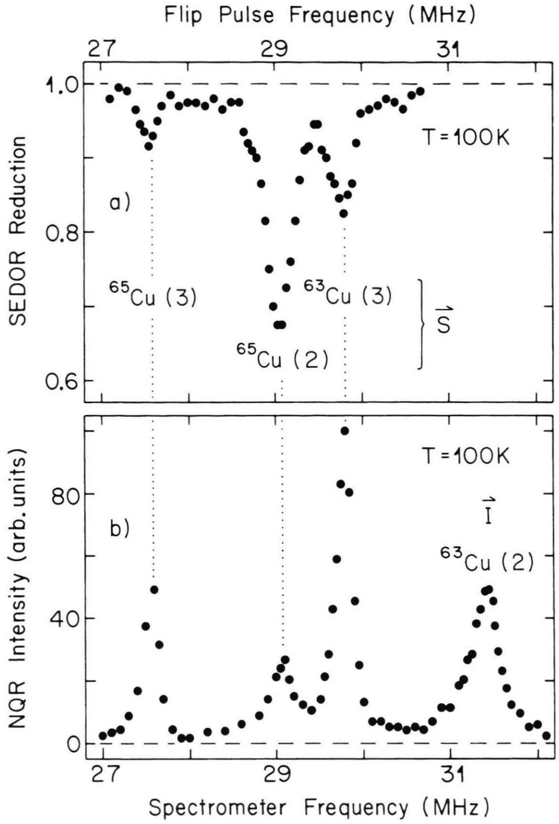

Fig. 5. SEDOR spectrum (a) and NQR spectrum (b) of planar Cu sites in $Y_2Ba_4Cu_7O_{15}$ [21]. The spin-echo of the ^{63}Cu(2) nucleus is observed. The vertical dotted lines indicate the NQR frequency of the S spin with which I interacts for a particular flip pulse frequency.

S at I. Hence the echo formation of I is disturbed, resulting in a reduction of the echo. We call this an $I-S$ SEDOR experiment. By comparing the I spin echo with and without the S spin flipping one can deduce the coupling between I and S spins. Details of the experiments are given in [21].

Figure 5 serves as an example. The lower part shows the planar Cu NQR spectrum of $Y_2Ba_4Cu_7O_{15}$ powder at 100 K. The upper part displays the reduction of the I spin-echo in an $I-S$ SEDOR with variable S spin flip frequency. In this case, I is the ^{63}Cu(2) nucleus and its echo reduction has been measured by sweeping the flip frequency from 27 to 30.5 MHz with both τ and τ_F kept constant. The I echo is reduced whenever the flip frequency coincides (as indicated by vertical dotted lines) with one of the NQR frequencies of the other signals. The SEDOR reduction thus reveals the coupling of the $^{63}Cu(2)$ nucleus with both $^{65}Cu(2)$ of the same plane and $^{63}Cu(3)$ and $^{65}Cu(3)$ of the adjacent inequivalent plane. The time dependence of the echo amplitude, $E_S(2\tau_F)$, can be described by a Gaussian:

$$E_S(2\tau_F) = E_{S0} \exp\left[-\frac{1}{2}\left(\frac{2\tau_F}{T_S}\right)^2\right], \quad (10)$$

where T_S denotes the SEDOR relaxatioin time.

Pennington et al. [6] showed that for *intra-plane* Cu SEDOR, T_S of the I spin is related to the S spin's time constant T_{2G} by

$$T_S^{\text{intra}}(I) = \frac{\gamma(S)}{\gamma(I)} T_{2G}(S), \quad (11)$$

where γ is the respective gyromagnetic ratio. T_{2G} is related to $T_{2G,\text{ind}}$ as discussed above.

By performing SEDOR experiments with various combinations of Cu nuclei within one plane and between both planes of a double plane we clearly demonstrated the existence of indirect spin-spin couplings in the plane and between planes. Since this coupling is mediated by exchange coupled electronic moments of planar Cu ions, the results are direct evidence for electronic inter-plane coupling.

In particular, we form the ratio of $T_{2G,\text{ind}}$ of the 1-2-4 block plane and $T_{S,\text{ind}}$ of the 63(2)–63(3) SEDOR. This ratio can be considered as the ratio of the inter-plane and intra-plane components of the *static* electron spin susceptibility and a such reflects the strength of the inter-plane coupling (represented by $T_{S,\text{ind}}^{-1}$) with respect to the intra-plane coupling (given by $T_{2G,\text{ind}}^{-1}$).

A first attempt to link $T_{2G,\text{ind}}/T_{S,\text{ind}}$ to the inter-plane exchange coupling constant, $J_\perp$, has been made by Millis and Monien [23]. Using a simiplified model for the susceptibility of two CuO_2 planes coupled by an inter-plane coupling $J_\perp$, the authors deduce from our experimental data the product of $J_\perp$ and the maximum value of the susceptibility, $\chi_{\max}$. At 120 K, they get for this product 0.4 which, together with $\chi_{\max} \approx 80$ states/eV–Cu [7], would imply $J_\perp \approx 5$ meV. Although small, this value appears to be of the right order of magnitude. Obviously, for a more reliable value, a more involved analysis is necessary.

6. Relation Between Spin Gap and Inter-plane Coupling

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 inter-plane effects.

Pursuing the first path, Sokol and Pines [24] propose a 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 $T_1 T$ and $T_{2G,\text{ind}}$ values. In the *quantum critical* (QC) regime (applicable to spin gap compounds), the ratio $T_1 T/T_{2G,\text{ind}}$ is constant while in the *overdamped* (OD) regime $T_1 T/T_{2G,\text{ind}}^2$ is constant.

We have checked these predictions [15] and found that $YBa_2Cu_3O_7$ is in the OD regime from T_c up to 300 K, and that the underdoped $YBa_2Cu_4O_8$ structure is in the QC regime for temperatures above T^*. However, $Y_2Ba_4Cu_7O_{15}$ does not fit into this scheme. Instead, taking the inter-plane coupling as the origin of the spin gap, quite naturally accounts for the observed $Y_2Ba_4Cu_7O_{15}$ behavior. The inter-plane coupling not only dictates a common temperature dependence of the dynamic susceptibility in adjacent planes, it also leads to a *common* value of the spin gap in the fluctuation spectrum. Alhough we cannot demonstrate that the inter-plane coupling is indeed responsible for the occurrence of the spin gap, our results point to the importance of the interplane coupling in the spin gap formation.

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