Nuclear Spin-lattice Relaxation in Cuprate Superconductors – Some New Approaches

Detlef Brinkmann[a] and Andrei Yu. Zavidonov[a,b,c]

[a] Physik-Institut, University of Zürich, CH-8057 Zürich, Switzerland
[b] Physics Department, Kazan State University, 420008 Kazan, Russia
[c] Present address: MATH POINT LTD, 509-720 Spadina Ave, Toronto, ON M5S 2H8, Canada

Reprint requests to Prof. D. B.; E-mail: Detlef.Brinkman@physik.unizh.ch

This paper reviews theoretical studies of nuclear spin-lattice relaxation we have made for the normal state of the cuprate high-temperature superconductors $YBa_2Cu_3O_7$, $YBa_2Cu_4O_8$, and $La_{2-x}Sr_xCuO_4$. In the case of *planar* sites, we calculated the dynamic spin susceptibility within a constraint-free theory based on the presentation of the $t - J$ model in terms of Hubbard operators. The results for ^{63}Cu, ^{17}O, and ^{89}Y are in good agreement with experimental data. The relaxation (and the Knight shift) of *chain* Cu in $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$ requires a different treatment; our approach uses the Luttinger-liquid model. Again, good agreement with experiment is achieved.

Key words: NMR Relaxation; $t - J$ Model; Cuprate Superconductors.

1. Introduction

NMR (Nuclear magnetic resonance) and NQR (Nuclear quadrupole resonance) are still playing an important role to understand high-temperature superconductors at the atomic level [1, 2]. In particular, studies of the various nuclear relaxation times are an essential and, quite often, indispensable tool to decipher the complex behavior of the electronic systems. Here, we will be concerned with nuclear *spin-lattice* relaxation in the *normal* state of cuprate superconductors and especially with those cases where the time-dependent perturbations can be expressed in terms of fluctuating fields arising from electronic spins. In this paper, we will review our calculation of relaxation times performed in recent years by using some new approaches [3 - 6].

Cuprate superconductors we are concerned with are the compounds $YBa_2Cu_3O_{7-\delta}$, $YBa_2Cu_4O_8$ and $La_{2-x}Sr_xCuO_4$ which are derived from antiferromagnetic (AF) parent compounds by doping (in the case of the Y structures) or substitution. Both processes create electron holes in the CuO_2 planes and destroy the AF long-range order, while AF short-range order is still present in the superconducting compounds. Hence, there are powerful fluctuations which can cause spin-lattice relaxation. The hole dynamics in the AF background is perfectly described by the dynamic spin susceptibility, $\chi(q, \omega)$, which depends on wave-vector, q, and frequency, ω, and thus provides direct information about the low energy excitation spectrum and its evolution with doping.

From $\chi(q, \omega)$, parameters like the spin-lattice relaxation rate, $1/T_1$, can be derived. This rate is given by the Moriya formula [7] which reads, if T_1 is measured by NQR,

$$\frac{1}{T_1} = \frac{2k_B T}{g^2 \mu_B^2 \hbar} \sum_q F(q) \frac{\chi''_{+-}(q, \omega_0)}{\omega_0}, \qquad (1)$$

where ω_0 is the NQR frequency. The form factors, $F(q)$, cause the differences in the temperature dependence of relaxation rates of different isotopes [8, 2] such as planar ^{63}Cu, planar ^{17}O and ^{89}Y; details will be discussed in Section 4.

Obviously, the quality of calculating $1/T_1$ is based on our knowledge of the imaginary part of the dynamic spin susceptibility, $\chi''_{+-}(q, \omega_0)$. Several models have been employed to calculate $1/T_1$ for the normal state of high-temperature superconductors;

this will briefly be discussed in Section 2 together with our alternate approach of calculating the dynamic susceptibility. In the remaining sections we will review our $1/T_1$ calculations for planar ^{63}Cu in $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{7-y}$ and for ^{63}Cu, ^{17}O, and ^{89}Y in $YBa_2Cu_3O_7$. The relaxation of *chain* Cu in $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$ requires a different treatment; our approach using the Luttinger-liquid model will be discussed in Section 5.

2. The Dynamic Spin Susceptibility

Usually, calculations of the dynamic spin susceptibility start from the $t - J$ model [9]. In the CuO_2 planes, a hole resides primarily on O sites in a square of O atoms and forms, by hybridization, a strong bond with the central Cu^{2+} ion, thus producing a local singlet which may move through the lattice of Cu^{2+} ions. t is the hopping energy of the holes and J is the strong repulsion between holes residing on the same square.

Starting from the $t - J$ model, the susceptibility is usually calculated by using various methods like the diagrammatic [10, 11], projection [12], slave-boson [13, 14] or slave-fermion [15, 16], and the extended Dyson representation method [17]. However, in spite of considerable progress, all theories have some disadvantages which are mainly connected with the use of either the mean-field approximation for the local constraints of operators or the random phase approximation (RPA). Therefore we calculated the dynamic spin susceptibility [3] within a constraint-free theory based on the presentation of the $t - J$ model in terms of Hubbard operators.

Our calculation goes as follows [3]. The spin part of the $t - J$ model can be modeled by a spin-1/2 Heisenberg antiferromagnet on a square lattice with a Hamiltonian

$$H_{t-J} = \sum_{i,j,\sigma} t_{ij} X_i^{\sigma 0} X_j^{0\sigma} + \sum_{i>j} J_{ij}(\mathbf{S}_i\mathbf{S}_j - \frac{1}{4}n_in_j). \quad (2)$$

Here, $\mathbf{S}_i$ are spin-1/2 operators at the lattice sites i, J_{ij} is a measure of the AF coupling between nearest neighbor sites i, j, and $X_i^{\sigma 0}$ are the Hubbard operators that create an electron with spin σ at site i. The hopping integral, t_{ij}, describes the motion of electrons without causing a change in their spins. The spin and density operators are defined as follows:

$$S_i^\sigma = X_i^{\sigma\overline{\sigma}}, S_i^z = \frac{1}{2}\sum_\sigma \sigma X_i^{\sigma\sigma}, n_i = \sum_\sigma X_i^{\sigma\sigma}; \sigma = -\overline{\sigma}$$

with the standard normalization $X_i^{00}+X_i^{++}+X_i^{--} = 1$. Without loss of generality, we can measure all energies from the center of gravity of the band.

When investigating the thermodynamic properties of the Heisenberg model by analytical methods, one must keep in mind that these methods are only valid at low temperatures if $T \ll 2\pi\rho_S$, where ρ_S is the stiffness. In doped cuprates, ρ_S becomes small and the above relation does not hold at the intermediate and high temperature range that is of most practical interest. We therefore employed the Green's function method [18] which is applicable at all temperatures; it naturally allows to comprise the hopping term since the method uses the Lee algebra for Hubbard operators. In this application, two points are worth mentioning. First, when evaluating some Hubbard commutators we did not reject a term which is responsible for the spin-spin correlations between Cu spins and, hence, becomes very important in the case of low-dimensional spin systems such as our two-dimensional (2D) system. Second, when dealing with the higher-order Green's function by using the Kondo and Yamaji [18] decoupling scheme, we kept the two parameter α and β, appearing in this scheme, different.

Our result for the dynamic spin susceptibility becomes

$$\chi_{+-}(\boldsymbol{q},\omega) = \frac{\omega\chi_0(\boldsymbol{q},\omega) + 4Jzc_1(1 - \gamma_q)Z(\boldsymbol{q},\omega)}{\omega\chi_1(\boldsymbol{q},\omega) + (\omega^2 - \omega_q^2)Z(\boldsymbol{q},\omega)}, \quad (3)$$

where

$$\chi_0(\boldsymbol{q},\omega) = \sum_k \frac{f_k^h - f_{k+q}^h}{\omega + E_k - E_{k+q}}, \quad (4)$$

$$\chi_1(\boldsymbol{q},\omega) = J_q\chi_0(\boldsymbol{q},\omega) + \sum_k \frac{\varepsilon_k f_k^h - \varepsilon_{k+q} f_{k+q}^h}{\omega + E_k - E_{k+q}}, \quad (5)$$

$$Z(\boldsymbol{q},\omega) = \sum_k \frac{1}{\omega + E_k - E_{k+q}}. \quad (6)$$

$\chi_0(\boldsymbol{q},\omega)$ denotes the dynamic susceptibility of free holes. $\chi_1(\boldsymbol{q},\omega)$ has a similar meaning as the exchange enhancement factor in the RPA, where, however, the second term of $\chi_1(\boldsymbol{q},\omega)$ is equal to 1. In our theory, this second term is due to the strong electron correlations and provides the correct concentration behavior of $\chi_{+-}(\boldsymbol{q},\omega)$ at half-filling ($\delta = 0$) [19]. $Z(\boldsymbol{q},\omega)$ is a convenient abbreviation.

The other quantities appearing in (3 - 6) have the following meaning. c_1, c_2: the nearest and next-nearest spin-spin correlation function, respectively, of Cu spins. z: number of nearest neighbors of spin l. $\gamma_q = (2/z) \sum_{\alpha=1}^{z/2} \cos q_\alpha$. E_k, ε_k: total and kinetic energy, respectively, of holes in the absence of correlations. $f_k^h(-E_k + \mu)$: Fermi function of holes with $\mu =$ chemical potential.

How reliable is our model? To answer the question, we have considered the case of the 2D Heisenberg antiferromagnet (with $z = 4$) where numerical results for the relevant parameters are already known. Since now $f_k^h = 0$, the low-energy excitations predicted by the $t - J$ model are spin waves with energies ω_q. In the limit of $T = 0$, the self-consistent equations for the parameters c_1, c_2, and α can be solved exactly.

We then found, among others: c_1, c_2, and χ_S agree remarkably well with those of a modified spin-wave theory; the χ_S value is compatible with results of the non-linear σ model and the isotropic spin-wave theory; χ_S, c_1, and the internal energy $u = 6c_1$ agree quite well with Monte Carlo data.

Having established the basic reliability of our model, we then evaluated the AF correlation length in a 2D Heisenberg antiferromagnet; details are given in [3]. Our model is able to reproduce the main features of the temperature and doping dependences of the correlation length in both the pure Heisenberg antiferromagnet (e.g. La_2CuO_4) and doped compounds (e.g. $La_{2-x}Sr_xCuO_4$).

3. Planar ^{63}Cu Relaxation in $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{7-y}$

We will now discuss the first application of the results described in Sect. 2: we will deal with the relaxation of planar copper in the normal state of the superconductors $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{7-y}$ [5].

In a first step, one has to calculate the one-particle excitation energy, E_k, of holes; we used the retarded Green's functions formalism. By δ we denote the number of extra holes, due to doping, per one plane Cu^{2+}. The result is

$$E_k = 2t_{eff}(\cos k_x + \cos k_y),\qquad(7)$$

where the effective hopping integral is

$$t_{eff} = t\left(p + \frac{2c_1}{p} - \frac{p^2}{2p-1}I^2 + \frac{J}{2t}I\right)\qquad(8)$$

with

$$I = \frac{1}{N}\sum_k \gamma_k f_k^h, \quad p = \frac{1+\delta}{2}.\qquad(9)$$

Equation (8) contrasts strongly with the Hubbard-I approach [20] which corresponds to setting $t_{eff} = tp$. Suppression of t_{eff} by the nearest-neighbor spin correlations, c_1, is easily understood because a hole when moving through the Cu lattice retains its spin orientation.

We like to stress that t_{eff} [see (8)] exhibits the correct doping dependence. We found that at high hole concentrations ($\delta \sim 1$), the width $W = 8t_{eff}$ of the conducting band E_k approaches, as it should be, the value $W^{nonint} = 8t$ for the non-interacting case [21]. In contrast, at low doping ($\delta < 0.05$), we found that $W^{low} \sim t$ and weakly varies with doping. Using $J/t = 0.3$, which is a reasonable value for the $t - J$ model [21], we obtain $W^{low} \sim 3J$. This result agrees well with Monte Carlo data based on the $t - J$ model [22].

We are now ready to calculate $1/T_1$ according to (1). It follows from (3), that $\chi_{+-}(q, \omega_0)$ is strongly peaked close to the AF wave vector $q = Q = (\pi, \pi)$. Therefore, we replace q by Q in $\chi_0(q, \omega_0)$ and $\chi_1(q, \omega_0)$ since these functions vary weakly with q near Q [3]. The term $Z(q, \omega_0)$, see (6), can be calculated exactly, with an accuracy of the order of ω_0^2. After some calculations, one obtains

$$\frac{1}{T_1} = \frac{64\delta(A_\perp - 4B)^2}{C(\xi)\hbar(1+\delta)}\left(\frac{2t_{eff}}{Jp} - 1\right)\frac{k_BT|c_1|}{J^2g_-^2}\xi^2, \quad(10)$$

where $\xi = 1/2\sqrt{g_+ - 1}$ is the AF correlation length measured in terms of the lattice period and $C(\xi) = 7/5 + 30\exp[-3(1/2\xi)^{1/4}]$ with the abbreviations $g_+ = (1 + 30c_2 + 4\alpha|c_1|)/g_-$ and $g_- = 16\alpha|c_1|$. $A_\perp$ and B are the Cu on-site and transferred hyperfine couplings, respectively.

Since $1/T_1$ is proportional to ξ^2, the short-range AF correlations between Cu spins have a strong influence on relaxation, even in doped samples. This result is consistent with the basic idea of the *nearly antiferromagnetic Fermi liquid* (NAFL) description of high-temperature superconductors [23], where the dynamic spin susceptibility is generally written, in a phenomenological approach, as the superposition of two terms, one for itinerant quasiparticles and the

other for localized Cu^{2+} magnetic moments. Likewise, we find the influence of the hole subsystem on $1/T_1$ to be important. As we shall see below, ξ is strongly reduced with doping thus leading to the decrease of the Cu relaxation rate.

Our model predicts a divergence of ξ at $T = 0$, in disagreement with experiment. Since, according to neutron scattering measurements in slightly doped $La_{2-x}Sr_xCuO_4$, ξ saturates at low temperatures, we replace ξ by an *effective* correlation length:

$$\xi_{\text{eff}}^{-1} = \xi_0^{-1} + \xi^{-1}. \quad (11)$$

This equation is different from Keimer's empirical formula [24] where ξ does not reflect an influence of the hole subsystem. However, in reality, especially for conducting samples, the short-range order is essentially modified by moving holes.

The parameter ξ_0 is the value of ξ_{eff} at $T = 0$. Despite many attempts to interpret ξ_0, the meaning of this parameter is still unclear because of lack of any exact analytical result. Therefore, in our theory, ξ_0 is a variational parameter whose value is obtained by comparison with experiment.

We now fit (10) to experimental data for $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_7$. The value of δ can be identified with x in the case of $La_{2-x}Sr_xCuO_4$. For $YBa_2Cu_3O_7$, we regard $\delta \approx 0.2$ as a reasonable choice according to photoemission data. The quantities c_1, c_2, and α were calculated. Because the temperature dependence of $1/T_1$ is mainly determined by ξ_{eff} and the factor k_BT, we take for $|c_1|$ and g_- their values at $T = 0$. The calculated c_1 values are as follows: $c_1(x = 0.075) = -0.0884$, $c_1(0.1) = -0.0780$, and $c_1(0.15) = -0.0577$ for $La_{2-x}Sr_xCuO_4$; $c_1(\delta = 0.2) = -0.0375$ for $YBa_2Cu_3O_7$.

Thus, there are four parameters to be determined: $A_\perp - 4B$, t, J, and ξ_0, all of which, except ξ_0, will be fixed by values known either from experiment or calculations. We use $A_\perp - 4B \approx 190$ and $140\,kOe/\mu_B$ for $La_{1.85}Sr_{0.15}CuO_4$ and $YBa_2Cu_3O_7$, respectively, [27] and $J = 0.1$ eV and $t = J/0.3$ [21]. Thus, ξ_0 is the only free parameter to be determined by fitting the experimental data. Figure 1 shows the result of the fit to the Cu relaxation rate. The high quality of the fit is taken as evidence for the reliability of our theory. The result is: $\xi_0(x = 0.075) = 14$, $\xi_0(0.1) = 9.5$, and $\xi_0(0.15) = 6.5$ for $La_{2-x}Sr_xCuO_4$ and $\xi_0(\delta = 0.2) = 6$ for $YBa_2Cu_3O_7$.

How does the correlation length ξ influence the relaxation rate? Figure 1 demonstrates that $1/T_1$ in
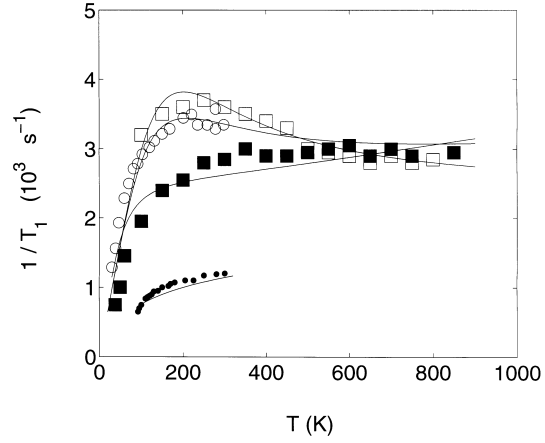

Fig. 1. Calculated temperature and doping dependence of the plane copper spin-lattice relaxation rate $1/T_1$ (solid lines) compared with experimental data for $La_{2-x}Sr_xCuO_4$ ([25]; open squares: $x = 0.075$, open circles: $x = 0.10$, full squares: $x = 0.15$) and $YBa_2Cu_3O_7$ ([26], full circles).

$La_{2-x}Sr_xCuO_4$ (for $x = 0.075, 0.1$) decreases with increasing temperature if the temperature is sufficiently high. Such a behavior is also found in the pure Heisenberg antiferromagnet where it is attributed to a specific temperature dependence of the AF correlation length, namely $\xi^{HA} \sim \exp(2\pi\rho_S/k_BT)$ [28]. However, in *doped* compounds, ξ^{HA} must be modified [3]:

$$\xi \sim \frac{Jp\sqrt{g_-}}{\sqrt{T(2t_{\text{eff}} - Jp)\delta}} \exp(2\pi\rho_S/k_BT), \quad (12)$$

where the stiffness ρ_S itself depends on doping. Our calculation [5] yields $2\pi\rho_S/k_B = 307$ K and 245 K for $x = 0.075$ and 0.1, respectively. For optimal doping, *i.e.* $x = 0.15$, however, $2\pi\rho_S/k_B$ is sufficiently small, namely 96 K, and in $YBa_2Cu_3O_7$, ρ_S is even negligible, so that the factor $T\xi_{\text{eff}}^2/C(\xi_{\text{eff}})$ in (10) *increases* with increasing temperature thus determining the increase of the Cu relaxation rate as observed experimentally.

In terms of the temperature dependence of $1/T_1$, our theory predicts two regimes which are governed by the temperature dependence of the AF correlation length ξ_{eff}. At low temperatures, when $\xi_0 \ll \xi$, $\xi_{\text{eff}} \simeq \xi_0$, $1/T_1$ displays a Korringa-like behavior. At high temperatures, ξ is much smaller than ξ_0 and $\xi_{\text{eff}} \simeq \xi$. Thus, according to (10) and (12), $1/T_1$ has a tendency to saturate at high temperatures:

$$\frac{1}{T_1^{\text{sat}}} \sim \frac{(A_\perp - 4B)^2 |c_1|}{\hbar J g_-} - O(T\xi^3/\xi_0). \quad (13)$$

These results agree with experiments.

Finally, the doping dependence of $1/T_1^{\text{sat}}$ is determined by the factor $|c_1|/g_-$. For $La_{2-x}Sr_xCuO_4$ with $x = 0.075$, 0.1, and 0.15, we get $|c_1|/g_- = 0.025$, 0.024, and 0.021, respectively. So, $1/T_1^{\text{sat}}$ in $La_{2-x}Sr_xCuO_4$ is almost doping independent at high temperatures, in agreement with experiment. In $YBa_2Cu_3O_7$, $|c_1|/g_- = 0.016$ is smaller than in $La_{2-x}Sr_xCuO_4$ because the spin-spin correlation function $|c_1|$ is rapidly reduced with doping in overdoped samples.

4. ^{63}Cu, ^{17}O, and ^{89}Y Relaxation in YBa$_2$Cu$_3$O$_7$

In this section, we will describe how we extended our previous results to the calculation of $1/T_1$ for plane ^{63}Cu and ^{17}O, and for ^{89}Y in the normal state of $YBa_2Cu_3O_7$ [6]. Experimental data for this special compound have revealed puzzling phenomena: the non-Korringa temperature dependence of $^{63}(1/T_1)$ and the nearly Korringa-like temperature dependence of $^{17}(1/T_1)$ and $^{89}(1/T_1)$ [2].

This contrasting behavior had motivated Millis, Monien, and Pines (MMP), to create the NAFL model (mentioned above) [23] which postulates both localized Cu^{+2} magnetic moments and free oxygen holes. Using the concept of hyperfine form factors [8], MMP found that AF fluctuations are dominating the ^{63}Cu relaxation while long-wave fluctuations caused by free holes are determining the ^{17}O and ^{89}Y relaxation. Recently, the MMP model has been quantitatively elaborated by Zha, Barzykin, and Pines (ZBP) [29] by taking into account incommensurate spin fluctuations and more general hyperfine coupling possibilities.

However, in spite of considerable progress, both theories, MMP and ZBP, have some disadvantages which are mainly connected with their phenomenological characters. For example, in these theories the temperature and doping dependence of the AF correlation length, ξ, which is the essential parameter that governs the temperature and doping dependence of $1/T_1$, are postulated or, at best, are taken from a comparison with experiment. Also, the MMP and ZBP expressions for the dynamic spin susceptibility are good approximations only for wave vectors in the vicinity of the AF wave vector. This implies that the MMP and ZBP theories provide reliable results

for $1/T_1$ if ξ is large. In $YBa_2Cu_3O_7$, however, ξ is small, as shown by neutron scattering measurements, and thus a more detailed theoretical analysis of NMR data in this compound is required.

As mentioned in the Introduction, the factor $F(q)\chi''_{+-}(q,\omega)/\omega$ is the essential function governing spin-lattice relaxation where the form factors determine the different temperature behavior of $1/T_1$ for different nuclei; the form factors filter the fluctuations at different points of the Brillouin zone. Thus we need to calculate the quantity $Q \equiv (F(q)/F(0))\chi''_{+-}(q,\omega)/\omega$ for Cu, O, and Y.

For the applied magnetic field along the c crystal axis, which is perpendicular to the CuO_2 planes, the $F(q)$'s are given by [2]

$$^{63}F_c(q) = (A_{ab} + 4B\gamma_q)^2, \quad ^{17}F_c(q) = 2C^2(1 + \gamma_q),$$

$$^{89}F_c(q) = 32D^2 \cos^2(q_x/2) \cos^2(q_y/2).$$

Here, A_{ab} is the on-site and C, D and B are the transferred hyperfine coupling constants. For C we adopted the formula [2] $C^2 = (C_\parallel^2 + C_\perp^2)/2$, where $C_\parallel$ and $C_\perp$ are the hyperfine couplings for two axes perpendicular to c.

Thus, it remains to calculate $\chi''_{+-}(q,,\omega_0)$ which can be written as

$$\chi''_{+-}(q,\omega_0) = \omega_0\chi_S[\Gamma_q^{-1}(0) + \Gamma_q^{-1}(AF)], \quad (14)$$

where

$$\Gamma_q^{-1}(0) = \frac{\Lambda_q}{1 - \gamma_q}, \quad \Gamma_q^{-1}(AF) = \frac{\Lambda_q(2g_+ + 1 + \gamma_q)}{(g_+ + \gamma_q)^2},$$

$$\Lambda_q = \frac{\chi_1'(q)[1 - \chi_S^{RPA}(q)/\chi_S(q)]}{J^2 g_-(g_+ + 1)^2 |Z(q)|}.$$

Here, $\chi_S = \chi_{+-}(0,0)/2$ and $\chi_S(q) = \chi_{+-}(q,0)/2$ are the static spin susceptibilities, and $\chi_S^{RPA}(q) = \chi_0'(q,0)/(2\chi_1'(q,0))$ is the static spin susceptibility calculated in the random phase approximation.

A numerical calculation of $|Z(q)|$ cannot be performed because of the presence of singularities in the denominator of (6). Instead, we calculated $|Z(q)|$ exactly, resulting in the expression

$$|Z(q)| = \frac{K(z)}{2\pi t_{\text{eff}}(|\sin(q_x/2)| + |\sin(q_y/2)|)},$$

$$z = \frac{2\sqrt{|\sin(q_x/2) \cdot \sin(q_y/2)|}}{|\sin(q_x/2)| + |\sin(q_y/2)|},$$

where $K(z)$ is a complete elliptic integral.

We calculated the response function $\chi''_{+-}(q,\omega)/\omega$ in the limit $\omega \to 0$ by using the parameters $J/t = 0.3$ and $\delta \approx 0.2$ (see Sect. 3), and the value $\xi_0 = 5$, which is derived from the best fit of $1/T_1$ to the experimental data (see below). The other parameters, which enter into $\chi''_{+-}(q,\omega)/\omega$, were calculated, except α, by using the self-consistent equations for the parameters c_1, c_2, and α (see Sect. 2). α can be derived from the exact diagonalization of the static spin susceptibility, χ_S^{ED}.

There are some important features of the response function $J^2\chi''_{+-}(q,\omega)/\omega$ in the limit $\omega \to 0$. For example, two kinds of excitations with $q \sim (0,0)$ and $q \sim (\pi,\pi)$ are dominating the low-energy excitation spectrum, consistent with the basic idea of the NAFL model. However, we find that the $q \sim (0,0)$ contribution in $\chi''_{+-}(q,\omega)/\omega$ cannot be interpreted as arising from free holes but rather from collective excitations including charge and spin degrees of freedom. Indeed, if the $q \sim (0,0)$ contribution would be due to free holes, the response $\chi''_{+-}(q \sim 0,\omega)/\omega$ would determine their static susceptibility, $\chi_0(0,0)$, that is $\chi_{+-}(q \sim 0,\omega)/\omega \sim \chi_0(0,0)$ [29]. In contrast, (14) tells us that $\chi''_{+-}(q \sim 0,\omega)/\omega$ is proportional to the static susceptibility, χ_S, of the *collective* excitations. Since χ_S cannot be represented in terms of independent contributions of hole and spin subsystems, one can conclude that the spin and charge degrees of freedom do not separate at $q \sim (0,0)$.

Our results for the quantity Q are plotted in [6]. For ^{63}Cu, ^{63}Q is large at the boundary $q \sim (\pi,\pi)$ of the Brillouin zone, thus making these nuclei sensitive to the AF correlations while ^{17}Q and ^{89}Q are zero. In contrast, ^{17}Q and ^{89}Q peak at the center, $q \sim (0,0)$, of the zone.

Using our Q values, we fit (1) to experimental data. (The factor 2 in the prefactor of the sum sign in this formula is now absent since we are dealing with NMR rather than NQR relaxation.) Five parameters are to be fitted: A_{ab}, B, C, D, and ξ_0. We handle ξ_0 as the only free parameter and fix the other parameters by using values known either from experiment or calculations: $A_{ab} + 4B \approx 203$ kOe/μ_B, $4B - A_{ab} \approx 141$ kOe/μ_B [27], $C \approx 22$ kOe/μ_B [30], and $D \approx 0.3$ kOe/μ_B [32].

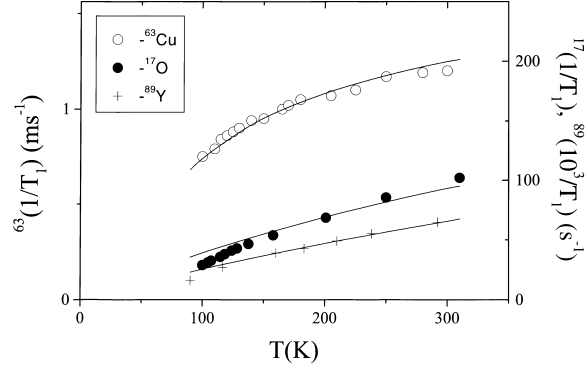The result of the fit is shown in Figure 2. The simultaneous fit to all three data sets yields $\xi_0 = 4.2$.

Fig. 2. Calculated temperature dependence of the relaxation rates $^{63}(1/T_1)$, $^{17}(1/T_1)$ and $^{89}(1/T_1)$ in YBa$_2$Cu$_3$O$_7$ (solid lines) compared with experimental data: Open circles from [31], filled circles and crosses from [30].

All three fits are good, which we take as evidence for the reliability of our theory. The value ξ_0 provides reasonable values for ξ_{eff}. For example, for $T_c = 90$ K and room temperature we found $\xi_{eff}(90\ \text{K}) = 1.73$ and $\xi_{eff}(300\ \text{K}) = 1.07$. Imai *et al.* [33] extracted ξ_{eff} from the temperature dependence of the copper nuclear spin-spin relaxation rate, $1/T_{2G}$ (G refers to Gaussian contribution), using the NAFL result $1/T_{2G} \sim \xi_{eff}$. They obtained $\xi_{eff}^{NMR}(90\ \text{K}) \approx 2.3$ and $\xi_{eff}^{NMR}(300K) \approx 1.5$ which is close to our result.

Figure 2 demonstrates that $^{89}(1/T_1)$ nearly follows the relation $^{89}(1/T_1) \sim T$ while $^{17}(1/T_1)$ exhibits a stronger deviation from this relation. The reason for the different behavior is that the form factor $^{17}F(q)$ does not suppress completely, in contrast to $^{89}F(q)$, the contribution of the spin fluctuations near $q \sim (\pi,\pi)$ to $\chi''_{+-}(q,\omega)$. Since $\chi''_{+-}(q \sim \pi,\omega)$ decreases with increasing temperature, one expects deviations from the relation $^{17}(1/T_1) \sim T$ at low temperatures. On the other hand, $^{63}(1/T_1)$ exhibits a very different temperature behavior. This implies that the contribution of the AF fluctuations to $\chi''_{+-}(q,\omega)$ for copper nuclei is dominating in the temperature range from 90 to 300 K.

5. Chain Cu Relaxation and Knight Shift in YBa$_2$Cu$_3$O$_7$ and YBa$_2$Cu$_4$O$_8$

To calculate the Cu spin-lattice relaxation in the Cu-O chains of cuprate superconductors requires a different treatment since these chains present an 1D quantum system. It is known that in 1D the Fermi-liquid paradigm, based on the quasi-particle picture,

breaks down and this then leads to the anomalies of the magnetic properties. If probed by NMR or NQR, chains do not exhibit simple metallic behavior as demonstrated, *e. g.*, by the Cu Knight shift [34] and $1/T_1$ [35 - 37].

It is believed that the Luttinger-liquid approach is most appropriate for the description of the properties of 1D quantum systems [38]. The notion of a "Luttinger liquid" was coined by Haldane [39] to describe the universal low-energy properties of 1D quantum systems, and to emphasize the principle difference between the Fermi-liquid and the Luttinger-liquid picture: the latter one takes care of interactions between the elementary excitations. Using the Luttinger liquid concept, some qualitative analysis of the temperature dependence of the nuclear spin-lattice relaxation in 1D systems has been done by Ren and Anderson [40] but only that contribution to $1/T_1$ has been considered which is due to the scattering processes with transfer momentum $\sim 2k_F$ (Kohn anomaly). However, as shown by NMR and NQR measurements on chains in YBa$_2$Cu$_3$O$_7$ and YBa$_2$Cu$_4$O$_8$, this contribution to $1/T_1$ is small, and thus a more detailed theoretical analysis of NMR and NQR data is required [4].

Our starting point for describing the charge and spin dynamics in the chains of cuprate superconductors is the 1D Hubbard model [41]. The low-energy properties in many 1D models, in particular the Hubbard model, can be described within the Luttinger-liquid approach [39]. Then, the Hubbard Hamiltonian simplifies and, in the continuum limit, reduces to the Luttinger-Tomonaga or, in other words, to the Gaussian model [40, 42]:

$$H_{LT} = \sum_{\nu=\rho,\sigma} \int_0^L dx \left[\frac{\pi v_\nu K_\nu}{2} \Pi_\nu^2 + \frac{v_\nu}{2\pi K_\nu}(\nabla \Phi_\nu)^2 \right].$$

$$(15)$$

The sum runs over all charge and spin degrees of freedom, ρ and σ, respectively, and the integration runs over a chain of length L. v_ρ and v_σ represent the charge and spin velocities, respectively. In the limit of large U ($t/U \ll 1$), where U is the value of the on-site Coulomb repulsion of electrons, these velocities can be calculated using the Bethe-ansatz [43]

$$v_\rho = 2t \sin(\pi c), \quad v_\sigma = \frac{J\pi}{2} \left[1 + \frac{\sin(2\pi c)}{2\pi(1-c)} \right], \quad (16)$$

where c is the concentration of *extra* holes, due to doping, per Cu(1) if one assumes that all copper in the chain are Cu^{2+}, and the exchange coupling constant, J, is given by $J = 4t^2/U$. The parameters K_ρ and K_σ describe the long-distance properties of the system. In particular, K_ρ determines the long-distance decay of all correlation functions of the Luttinger-Tomonaga model.

Details of the calculations are given in [4]. The result shows that there are two channels of magnetic relaxation, R_0 and R_{2k_F}, induced by quasiparticles with wave vectors $q \sim 0$ and $q \sim 2k_F$, respectively. So, the total relaxation rate is

$$\left(\frac{1}{T_1} \right)_{NQR} = R_0 + R_{2k_F}, \quad (17)$$

with

$$R_0 = \frac{3\pi(A + 2B)^2 \gamma_N^2 T}{4} \chi_S^2, \quad (18)$$

$$R_{2k_F} = \frac{3(A + 2B \cos 2k_F)^2 \gamma_N^2}{\pi^2 v_\sigma} \left(\frac{T}{v_\rho} \right)^{K_\rho} \Lambda_{\alpha\rho}, \quad (19)$$

where

$$\Lambda_{\alpha\rho} = \lim_{\alpha \to 0} \left(\frac{2}{\alpha\pi(K_\rho + 1)} \right)^{1-K_\rho} \frac{\pi}{\sin(\pi K_\rho/2)\Gamma(K_\rho)}.$$

Here, $\Gamma(z)$ is the gamma function. A is the on-site hyperfine field and B is the transferred field produced by the nearest neighbor Cu spins. χ_S is the static spin susceptibility. Further details may be found in [4].

We like to stress that our calculation, for this 1D system, also yields the spin part components of the magnetic shift tensor:

$$K^\zeta = K_{orb}^\zeta + \frac{A_\zeta + 2B}{\gamma_N \gamma_e} \chi_S, \quad (20)$$

where ζ denotes the crystal axes a, b, c with b lying along the chains and c perpendicular to the CuO$_2$ planes; K_{orb}^ζ is the orbital contribution to the total shift. NMR and NQR experiments [34, 35] show that for both, YBa$_2$Cu$_3$O$_7$ and YBa$_2$Cu$_4$O$_8$, the hyperfine fields A_ζ are almost isotropic. We replace A_ζ by the single value A.

We now will fit our expressions for the Cu magnetic shift and relaxation rate to experimental data. There are seven parameters entering the equations to be fitted: $A, B, U, t, c, T_0,$ and K_{orb}^c, where $2T_0$
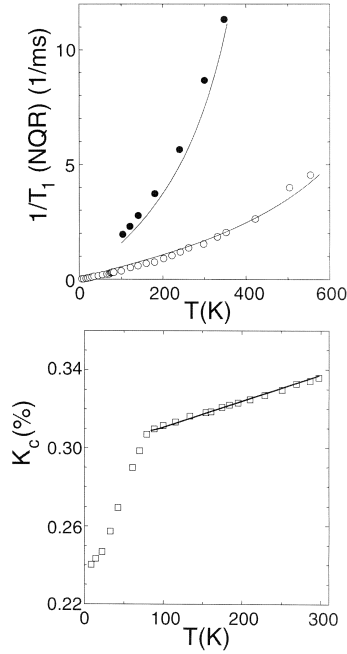
Fig. 3. *Top:* The calculated temperature dependences of the chain Cu spin–lattice relaxation rate, $(1/T_1)_{NQR}$ (full lines), fitted to experimental data for $YBa_2Cu_3O_7$ (closed circles, from [35]) and $YBa_2Cu_4O_8$ (open circles, from [36]). *Bottom:* The calculated temperature dependence of the component K^c of the chain Cu magnetic shift tensor (full line) fitted to experimental data for $YBa_2Cu_4O_8$ (open squares, from [34].

is identified as the band width energy cut-off, E_0. We will fix the parameters A, B, U, t and c by using values known either from experiment or calculations. In the chains of $YBa_2Cu_3O_7$, the on-site hyperfine field is $A \approx 30$ kOe/μ_B, while the transferred field is $B \approx 55$ kOe/μ_B [8]. For the hopping integral, t, and the one-site Coulomb repulsion, U, we use the values $t = 0.43$ eV, $U = 5.4$ eV which are valid for the plane [44]. Given the many structural similarities between the chains in $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$, we used the same parameters A, B, U, t for both compounds. The c values are known from photoemission experiments with chains: $c \approx 0.6$ for $YBa_2Cu_3O_7$ [45] and $c \approx 0.23$ for $YBa_2Cu_4O_8$ [46]. Because of the double chains, each Cu in $YBa_2Cu_4O_8$ has four nearest copper neighbors, hence the transferred coupling contains two contributions: a contribution B from copper sites within the same chain and a second one, which is approximately $-B/3$ [47] from copper sites of the nearest chain. Therefore, the

transferred field B in $YBa_2Cu_4O_8$ should be replaced by $2B/3$.

The fit of (17) and (20) to the respective experimental data is given in Fig. 3 with K^c_{orb} and E_0 as the only free parameters. (We did not consider the temperature dependence of the Knight shift in $YBa_2Cu_3O_7$ chains because they are controversial.) All three fits are very satisfactory, which we take as evidence for the reliability of the Luttinger-liquid picture. The best fit yields the following parameters: $E_0 = 1800$ K for $YBa_2Cu_3O_7$ and $E_0 = 3200$ K, $K^c_{orb} = 0.125\%$ for $YBa_2Cu_4O_8$. Using now our parameters and (20), we found a value $K^c(T = 100) = 0.41\%$ for the $YBa_2Cu_3O_7$ chains. This result is close to the experimental value of $K^c_{exp}(100) = 0.334 \pm 0.01\%$ [35].

A problem remains for $YBa_2Cu_4O_8$. According to Fig. 3 Bottom, the experimental value of the orbital shift at $T = 0$ is $K^{exp}_{orb}(T = 0) \approx 0.24\%$, if we assume the spin part of the Knight shift to be completely suppressed due to proximity-induced superconductivity in the CuO chains. This value disagrees with our fit result, $K^c_{orb} = 0.125\%$. On the other hand, $(1/T_1)_{NQR}$ below T_c is only slightly affected by the on–set of superconductivity (see Fig. 3 top). We see two possible explanations: (i) some fraction of the spin excitations is not suppressed by superconductivity and, hence, these excitations will provide a *finite* spin contribution to the Knight shift at $T = 0$; (ii) at temperatures below T_c, the interchain interaction becomes important and the Luttinger-liquid description breaks down. More experiments are needed to clarify this problem.

6. Summary

We have presented some new approaches to determine the nuclear spin-lattice relaxation time, T_1, in the normal state of cuprate superconductors. One starting point is the calculation of the dynamic spin susceptibility within a constraint-free theory based on the presentation of the $t - J$ model in terms of Hubbard operators. This treatment yields results which allow one to reproduce the main features of the temperature and doping dependences of the AF correlation length in both the pure Heisenberg antiferromagnet and doped compounds.

We then calculated the temperature and concentration dependence of $1/T_1$ of the *plane* copper nuclei in $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_{7-y}$. The predictions fit the experimental data very well, thus yielding reasonable values for parameters such as the spin-spin

correlation function, the antiferromagnetic correlation length at 0 K, and the width of the conducting band and their dependence on doping.

The calculations were extended to determine the temperature dependencies of $1/T_1$ of ^{63}Cu, ^{17}O and ^{89}Y in $YBa_2Cu_3O_7$. The predictions fit the experimental data very well, thus yielding reasonable values for parameters such as the antiferromagnetic correlation length at 0 K.

Finally, we calculated, based on the hypothesis of a Luttinger liquid ground state within the 1D Hubbard model, the temperature and concentration dependences of $1/T_1$ and the Knight shift of the *chain* Cu nuclei in $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$. The experimental results are well fitted by the predictions of the Luttinger-Tomonaga model and the renormalization group theory. The fit yields parameters which are reasonable for both compounds.

[1] D. Brinkmann and M. Mali, in NMR – Basic Principles and Progress, ed. by P. Diehl, E. Fluck, H. Günther, R. Kosfeld, J. Seelig, vol. 31, p. 171 (Springer, Berlin 1994).

[2] A. Rigamonti, F. Borsa, and P. Carretta, Rep. Prog. Phys. **61**, 1367 (1998).

[3] A. Yu. Zavidonov and D. Brinkmann, Phys. Rev. B **58**, 12486 (1998).

[4] A. Yu. Zavidonov and D. Brinkmann, Phys. Rev. B Brief Reports **61**, 3282 (2000).

[5] A. Yu. Zavidonov, I. A. Larionov, and D. Brinkmann, Phys. Rev. B **61**, 15462 (2000).

[6] A. Yu. Zavidonov and D. Brinkmann, Phys. Rev. B **63**, 132506-1 (2001).

[7] T. Moriya, Spin Fluctuations in Itinerant Electron Magnetism, Springer, Berlin 1985.

[8] F. Mila and T.M. Rice, Physica C **157**, 561 (1989).

[9] F. C. Zhang and T. M. Rice, Phys. Rev. B **37**, 3759 (1988).

[10] Yu. A. Izyumov and J. A. Henderson, Int. J. Mod. Phys. B **8**, 1877 (1994).

[11] F. Onufrieva and J. Rossat-Mignod, Phys. Rev. B **52**, 7572 (1995).

[12] K. W. Becker and U. Muschelknautz, Phys. Rev. B **48**, 13 826 (1993).

[13] T. Tanamoto, H. Kohno, and H. Fukuyama, J. Phys. Soc. Japan **62**, 1455 (1993).

[14] Z. Y. Weng, D. N. Sheng, and C. S. Ting, Phys. Rev. B **52**, 637 (1995).

[15] C. L. Kane, P. A. Lee, T. K. Ng, B. Chakraborty, and N. Read, Phys. Rev. B **41**, 2653 (1990).

[16] A. Auerbach and B. E. Larson, Phys. Rev. B **43**, 7800 (1991).

[17] F. P. Onufrieva, V. P. Kushnir, and B. P. Toperverg, Phys. Rev. B **50**, 12 935 (1994).

[18] J. Kondo and K. Yamaji, Prog. Theor. Phys. **47**, 807 (1972).

[19] J. Hubbard and K. P. Jain, J. Phys. C (Pros. Phys. Soc.) **1**, 1650 (1968).

[20] J. Hubbard, Proc. Roy. Soc. London, Ser. A **276**, 238 (1963).

[21] E. Dagotto, Rev. Mod. Phys. **66**, 763 (1994).

[22] A. Moreo, S. Haas, A. W. Sandvik, and E. Dagotto, Phys. Rev. B **51**, 12 045 (1995).

[23] A. Millis, H. Monien, and D. Pines, Phys. Rev. B **42**, 167 (1990).

[24] B. Keimer, N. Belk, R. J. Birgeneau, A. Cassanho, C. Y. Chen, M. Greven, M. A. Kastner, A. Aharony, Y. Endoh, R. W. Erwin, and G. Shirane, Phys. Rev. B **46**, 14 034 (1992).

[25] T. Imai, C. P. Slichter, K. Yoshimura, and K. Kosuge, Phys. Rev. Lett. **70**, 1002 (1993).

[26] S. Ohsugi, Y. Kitaoka, K. Ishida, G.-q. Zheng, and K. Asayama, J. Phys. Soc. Japan **63**, 700 (1994).

[27] Y. Zha, V. Barzykin, and D. Pines, Phys. Rev. B **54**, 7561 (1996), p. 7566/7.

[28] S. Chakravarty, B. I. Halperin, and D. R. Nelson, Phys. Rev. B **39**, 2344 (1989).

[29] Y. Zha, V. Barzykin, and D. Pines, Phys. Rev. B **54**, 7561 (1996).

[30] V. A. Nandor, J. A. Martindale, R. W. Groves, O. M. Vyaselev, and C. H. Pennington, Phys. Rev. B **60**, 6907 (1999).

[31] S. E. Barrett, J. A. Martindale, D. J. Durand, C. H. Pennington, C. P. Slichter, T. A. Friedmann, J. P. Rice, and D. M. Ginsberg, Phys. Rev. Lett. **66**, 108 (1991).

[32] M. Takigawa, W. L. Hults, and J. L. Smith, Phys. Rev. Lett. **71**, 2650 (1993).

[33] T. Imai, C. P. Slichter, A. P. Paulikas, and B. Veal, Phys. Rev. B **47**, 9158 (1993).

[34] M. Bankay, M. Mali, J. Roos, and D. Brinkmann, Phys. Rev. B. **50**, 6416 (1994).

[35] C. H. Pennington, D. J. Durand, C. P. Slichter, J. P. Rice, E. D. Bukowski, and D. M. Ginsberg, Phys. Rev. B **39**, 2902 (1989).

[36] H. Zimmermann, M. Mali, D. Brinkmann, J. Karpinski, E. Kaldis, and S. Rusiecki, Physica C **159**, 681 (1989).

[37] F. Raffa, M. Mali, A. Suter, A.Yu. Zavidonov, J. Roos, D. Brinkmann, and K. Conder, Phys. Rev. B **60**, 3636 (1999).

[38] V. J. Emery, in Highly Conducting One Dimensional Solids, edited by J. T. Devreese et al., Plenum Press, New York 1978.

[39] F. D. M. Haldane, J. Phys. C **14**, 2585 (1981).

[40] Y. Ren and P. W. Anderson, Phys. Rev. B **48**, 16 662 (1993).

[41] P. W. Anderson, Science **235**, 1196 (1987).

[42] H. J. Schulz, Phys. Rev. Lett. **64**, 2831 (1990).

[43] H. Shiba, Phys. Rev. B **6**, 930 (1972).

[44] M. S. Hybertsen, M. Schlüter, and N. E. Christensen, Phys. Rev. B **39**, 9028 (1989).

[45] N. Nücker, E. Pellegrin, P. Schweiss, J. Fink, S. L. Molodtsov, C. T. Simmons, G. Kaindl, W. Frentrup, A. Erb, and G. Müller-Vogt, Phys. Rev. B **51**, 8529 (1995).

[46] A. Krol, Z. H. Ming, Y. H. Kao, N. Nücker, G. Roth, J. Fink, G. C. Smith, K. T. Park, J. Yu, A. J. Freeman, A. Erband, G. Müller-Vogt, J. Karpinski, E. Kaldis, and K. Schönmann, Phys. Rev. B **45**, 2581 (1992).

[47] P. Freund, J. Phys. C **16**, 5039 (1983).