Magnetic properties of chains in cuprate superconductors studied by the Luttinger-liquid model

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Mapping the one-dimensional Hubbard model into the Luttinger-Tomonaga model, we have calculated the temperature dependencies of the chain copper nuclear magnetic spin-lattice relaxation rate and Knight shift for the normal state of the superconducting materials $YBa_2Cu_3O_7$ and $YBa_2Cu_4O_8$. The dynamic spin susceptibility has been obtained by using a bosonization technique and results of the renormalization group analysis for one-dimensional quantum systems. A comparison of our results with experiment shows that the model is able to reproduce the main features of the spin dynamics in both materials.

One of the essential structural elements of high-temperature superconductor (HTSC) compounds such as $YBa_2Cu_3O_7$ (Y123) and $YBa_2Cu_4O_8$ (Y124), are single and double Cu-O chains, respectively. However, in spite of intensive experimental and theoretical studies, no established consensus has been reached for the chains' ground state and their low energy excitation spectrum.[1–3] The Cu-O chains present a good example of an one-dimensional (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 nuclear magnetic resonance (NMR) or nuclear quadrupole resonance (NQR), chains do not exhibit simple metallic behavior as demonstrated, e.g., by the Cu Knight shift[4] and the spin-lattice relaxation rate, $1/T_1$.[5–7]

It is believed that the Luttinger-liquid approach is most appropriate for the description of the properties of 1D quantum systems.[8] The notion of a ''Luttinger liquid'' was coined by Haldane[9] to describe the universal low-energy properties of 1D quantum systems, and to emphasize the principle difference between Luttinger-liquid and Fermi-liquid pictures. 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,[10] 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 Y123 and Y124, this contribution to $1/T_1$ is small and thus a more detailed theoretical analysis of NMR and NQR data is required.

In this Brief Report, we present, based on the Luttinger-Tomonaga model, the theory of the Cu magnetic spin-lattice relaxation and the Knight shift at the chain copper sites, Cu(1), in Y123 and Y124. Using the bosonization technique, we calculate the spin correlation functions and the dynamic spin susceptibility. The temperature dependence of the dynamic spin susceptibility is evaluated on the basis of the scaling theory for the 1D interacting electron gas.

Our starting point for describing the charge and spin dynamics in the chains of HTSC is the 1D Hubbard model[11] with its Hamiltonian

$$H = t \sum_{\langle ij \rangle, \sigma} c_{i\sigma}^{+} c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow}. \qquad (1)$$

Here, $c_{i\sigma}^{+}$ is the fermionic operator that creates an electron, with spin σ, at site *i*. t is the hopping integral and U is the value of the one-site Coulomb repulsion of electrons. Haldane[9] realized that the low-energy properties in many 1D models, in particular the Hubbard model, can be described within the Luttinger-liquid approach. Then, Eq. (1) simplifies and, in the continuum limit, reduces to the Luttinger-Tomonaga or, in other words, to the Gaussian model[10,12]

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

The sum runs over all charge and spin degrees of freedom, ρ and σ. We did not include the umklapp scattering term which is ineffective when the band is not half-filled, and we have taken into account that the backward scattering term is renormalized to zero in the long-wavelength limit.[8]

In Eq. (2), 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 ($t/U \ll 1$), which is realized in HTSC,[13] these velocities can be calculated using the Bethe ansatz[14]

$$v_\rho = 2t \sin(\pi c), \quad v_\sigma = \frac{J\pi}{2} \left[1 + \frac{\sin(2\pi c)}{2\pi(1-c)} \right], \qquad (3)$$

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+},[15] and the exchange coupling constant, J, is given by $J = 4t^2/U$. The parameters K_ρ and K_σ in Eq. (2) 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. At first order in t/U, the parameter K_ρ takes the value[16]

$$K_\rho = \frac{1}{2} - \frac{4t \ln 2}{\pi U} \cos \frac{\pi c}{2},$$

and spin-rotational invariance requires $K_\sigma = 1$.[17]

The phase fields in Eq. (2) are defined as[9]

$$\Pi_\nu(x) = \frac{1}{L} \sum_{p \neq 0} e^{-(\alpha |p| x/2 + ipx)} [\nu_{p+} - \nu_{p-}] + \frac{J_\nu}{L}, \qquad (4)$$

$$\Phi_\nu(x)=\frac{\pi}{iL}\sum_{p\neq 0}\frac{e^{-(\alpha|p|x/2+ipx)}}{p}[\nu_{p+}+\nu_{p-}]-N_\nu\frac{\pi x}{L}, \tag{5}$$

where J_ν is the current operator and N_ν and $\tilde{N}_\nu$ are number operators. In the thermodynamical limit, $L\to\infty$, these operators do not contribute to correlation functions.[17]

It is convenient, for later purposes, to define the phase field[9]

$$\Theta_\nu(x)=\frac{i\pi}{L}\sum_{p\neq 0}\frac{e^{-(\alpha|p|x/2+ipx)}}{p}[\nu_{p+}-\nu_{p-}]+\tilde{N}_\nu\frac{\pi x}{L}. \tag{6}$$

In Eqs. (4)–(6), the limit $\alpha\to 0$ should be taken.[18]

The Fourier components of the charge- and spin-density operators for the right- $(r=+)$ and left- $(r=-)$ going fermions, $\rho_r(p)$ and $\sigma_r(p)$, respectively, obey the Bose-like commutation relations: $[\rho_{pr},\rho_{p'r'}]=[\sigma_{pr},\sigma_{p'r'}]=-\delta_{r,r'}\delta_{p,p'}(rpL/2\pi)$.

Now, we calculate the copper Knight shift and relaxation rate. The hyperfine coupling of the Cu nuclei, with spin I, to the neighboring Cu electron spins is given by the Mila-Rice Hamiltonian[19]

$$H=\sum_\alpha\left[I^\alpha\left(A^\alpha S_0^\alpha+B\sum_i S_i^\alpha\right)\right], \tag{7}$$

where S_0 is the spin at the same nuclear site and A is the on-site hyperfine field; the nearest neighbor Cu spins, S_i, produce the transferred field, B.

In case of an 1D system, Eq. (7) leads to the following formula for the spin part components of the magnetic shift tensor:

$$K^\zeta=K^\zeta_{\rm orb}+\frac{A_\zeta+2B}{\gamma_N\gamma_e}\chi_S. \tag{8}$$

Here, χ_S is the static spin susceptibility and ζ denotes the crystal axes a, b, c with b lying along the chains and c perpendicular to the CuO_2 planes; K^ζ_{orb} is the orbital contribution to the total shift. NMR and NQR experiments[4,5] show that for both Y123 and Y124 compounds, the hyperfine fields A_ζ are almost isotropic and we replace A_ζ by the single value A.

The static susceptibility, χ_S, is derived in a standard way. Adding the Zeeman term to the Hamiltonian H_{LT} and taking the second derivative of the ground state energy with respect to the magnetic field, we have $(\mu_B=1)$

$$\chi_S=\frac{2}{\pi v_\sigma}, \tag{9}$$

which agrees with the exact result of Shiba[14] for the 1D Hubbard model.

The spin-lattice relaxation rate of the Cu(1) nuclei as measured by NQR, $(1/T_1)_{\rm NQR}$, is given by the Moriya formula:

$$\left(\frac{1}{T_1}\right)_{\rm NQR}=3\gamma_N^2T\sum_{\mathbf{q}}(A+2B\cos q)^2\frac{\chi''(\mathbf{q},\omega_Q)}{\omega_Q}, \tag{10}$$

$$\chi''(\mathbf{q},\omega_Q)=\mathrm{Im}\left(i\int_0^\infty dt e^{i\omega_Q t}\langle[S_q^-(t),S_{-q}^+(0)]\rangle\right). \tag{11}$$

Here, $\langle\cdots\rangle$ denotes the thermodynamical average, ω_Q is the NQR frequency, and S_q^+, S_{-q}^- represent the Fourier components of the spin one-half operators which, in the continuum limit, are $S_q^\pm=(1/\sqrt{L})\int_0^L dx e^{iqx}S^\pm(x)$.

To calculate the spin correlation function in Eq. (11), we use the pseudofermionic representation of the spin operators:

$$S^+(x)=\psi_\uparrow^+(x)\psi_\downarrow(x),\quad S^-(x)=(S^+(x))^*. \tag{12}$$

$\psi^+(x),\psi(x)$ are the fundamental fermionic operators which are related to the operators of the right- and left-going fermions as[9]

$$\psi_s(x)=\frac{1}{L}\sum_{kr}\theta(kr)\int_0^L dx' e^{ik(x-x')}\psi_{rs}(x'). \tag{13}$$

The remaining task is to construct the representation of the fermionic operators, ψ_{rs}, in the basis of the bosonic operators. This has been accomplished by Luther and Peschel;[18] we are using the more precise formulation by Haldane:[9]

$$\psi_{rs}(x)=\frac{e^{ixr(k_F-\pi/L)}}{\sqrt{2\pi\alpha}}U_{rs}^+\exp[-\Delta_{rs}(x)], \tag{14}$$

$$\Delta_{rs}(x)=\frac{i}{\sqrt{2}}[r\Phi_\rho(x)-\Theta_\rho(x)+s(r\Phi_\sigma(x)-\Theta_\sigma(x))].$$

Here, U_{rs}^+ is the ladder operator which increases by unity the number of fermions with the projection of spin s on branch r; s stands for $s=(+,-)\to(\uparrow,\downarrow)$, and $k_F=\pi(1-c)/2$ is the Fermi wave vector. Equations (12)–(14) tell us that the spin correlation function $\langle[S_q^-(t),S_{-q}^+]\rangle$ in Eq. (11) represents the thermal average of exponentials of linear combinations of bosonic operators. It is a major advantage of the bosonic representation that such expressions can be evaluated quite simply using the relation

$$\langle e^C e^D\rangle=e^{\langle(C+D)^2\rangle/2}e^{[C,D]/2}, \tag{15}$$

which is valid for a form linear in boson operators whose exponential is averaged with the harmonic oscillator Gaussian Hamiltonian of Eq. (2). Then, diagonalizing the Hamiltonian H_{LT} by the Bogoliubov transformation[8,17] yields $e^S H_{LT}e^{-S}\to\Sigma_{p,\nu}v_\nu|p|\nu(p)\nu(-p)$, $e^S\Phi_\nu e^{-S}\to\Phi_\nu\sqrt{K_\nu}$, and $e^S\Theta_\nu e^{-S}\to\Theta_\nu/\sqrt{K_\nu}$.

Using Eqs. (12)–(15), we finally get

$$\left(\frac{1}{T_1}\right)_{\rm NQR}=R_0+R_{2k_F}, \tag{16}$$

with the two contributions

$$R_0=\frac{3\pi(A+2B)^2\gamma_N^2T}{4}\chi_S^2, \tag{17}$$

$$R_{2k_F}=\frac{3(A+2B\cos 2k_F)^2\gamma_N^2}{\pi^2 v_\sigma}\left(\frac{T}{v_\rho}\right)^{K_\rho}\Lambda_{\alpha\rho}, \tag{18}$$

with

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

where $\Gamma(z)$ is the gamma function.

There are two channels of magnetic relaxation, R_0 and R_{2k_F}, induced by quasiparticles with wave vectors $\mathbf{q}\sim 0$ and $\mathbf{q}\sim 2k_F$, respectively. In the limit $\alpha\to 0$, Eq. (18) diverges. Such a singularity reflects very large momentum excitations in the Luttinger liquid, which do not occur in a real system with finite bandwidth.[18] Therefore, following Luther and Peschel,[18] we replace α by the finite value $\sim 1/p_{\max}\sim 1/\pi$, where $p_{\max}$ is the momentum cutoff for finite bandwidth. The contribution R_{2k_F} is proportional to T^{K_ρ} and thus agrees with results of Ren and Anderson.[10] Our estimations show that, for both Y123 and Y124, R_{2k_F} is much smaller than R_0 for all temperatures above T_c.

Our theory is not complete yet since the Gaussian model we used predicts a temperature *independent* behavior of χ_S that disagrees with the Monte Carlo results[20] for the 1D Hubbard model. We can improve our theory using the results of the renormalization group analysis for the 1D Hubbard model.[21] In first order, the temperature dependent screening of backward scattering is given by the expression[21] $g(T)=U/[1+U\ln(E_0/2T)/\pi v_\sigma]$. Then, the static susceptibility, $\chi_S^{\rm Hub}$, can be calculated using the random phase approximation:[22]

$$\chi_S^{\rm Hub}=\chi_S[1-g(T)/2\pi v_\sigma]^{-1}. \tag{19}$$

In all equations above, we now replace χ_S by $\chi_S^{\rm Hub}$.

Equation (19) can also be reproduced by other methods. For example, Ogata and Shiba[23] showed that for the 1D Hubbard model in the limit of large U, the Bethe-Ansatz function can be written as a product of a Slater determinant of spinless fermions and the spin wave function of the 1D $S=1/2$ Heisenberg model. This implies that the temperature behavior of $\chi_S^{\rm Hub}$ should be qualitatively similar to that of the Heisenberg model susceptibility. Using the conformal field theory for 1D $S=1/2$ Heisenberg antiferromagnet, one finds[24]

$$\chi_S^{\rm Heis}=\frac{2}{\pi v_\sigma}\left(1+\frac{1}{2\ln(T_0/T)}\right)+O((\ln T)^{-3})$$

that perfectly agrees with our result for $\chi_S^{\rm Hub}$ in the case of low temperatures, that is for $\ln E_0/2T\gg 1$, if one identifies T_0 as $E_0/2$. The parameter E_0 is the band width energy cut-off. It acts as the ultraviolet regulator of the perturbation theory for the weakly interacting electron gas. In the limit of large U, however, the meaning of E_0 is unclear because of lack of any exact analytical results in this limit. Therefore, in our theory E_0 is a variational parameter whose value is obtained by comparison with experiment.

We now will fit our expressions for the Cu magnetic shift [Eq. (8)] and relaxation rate [Eq. (16)] to experimental data. There are seven parameters entering the equations to be fitted: A,B,U,t,c,T_0 and $K^c_{\rm orb}$. We will fix the parameters A,B,U,t, and c by using values known either from experiment or calculations. In the chains of Y123, the one-site hyperfine field is $A\approx 30$ kOe/μ_B, while the transferred field is $B\approx 55$ kOe/μ_B.[19] 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.[25] Given the many structural similarities between the chains in Y123 and Y124, 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 Y123 (Ref. 26) and $c\approx 0.23$ for Y124 (Ref. 27). Because of the double chains, each Cu in Y124 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 $-B/3$,[28] from copper sites of the nearest chain. Therefore, the transferred field B in Y124 should be replaced by $2B/3$.

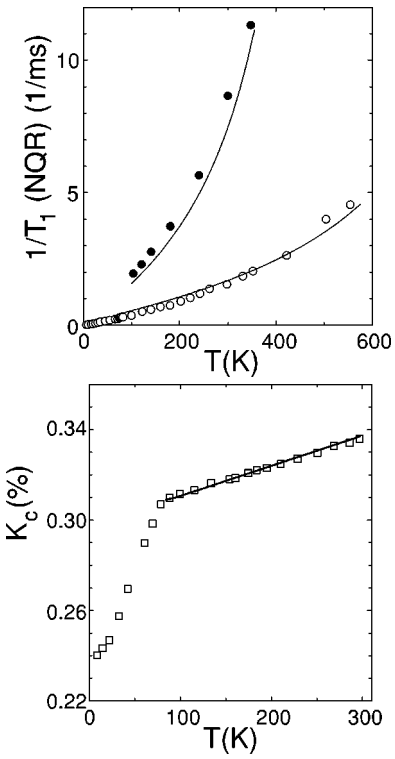

FIG. 1. Top: The calculated temperature dependences of the chain Cu spin-lattice relaxation rate, $(1/T_1)_{\rm NQR}$ (full lines), fitted to experimental data for Y123 (closed circles, from Ref. 5) and Y124 (open circles, from Ref. 6). Bottom: The calculated temperature dependence of the component K^c of the chain Cu magnetic shift tensor (full line) fitted to experimental data for Y124 (open squares, from Ref. 4).

Accepting this point of view, we have fitted Eqs. (8) and (16), with T_0 and $K^c_{\rm orb}$ as the only free parameters, to the experimental NQR relaxation rate of both Y123 and Y124 and to the Knight shift in Y124 (Fig. 1). We did not consider the temperature dependence of the Knight shift in Y123 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 Y123 and $E_0=3200$ K, $K^c_{\rm orb}=0.125\%$ for Y124. Using now our parameters and Eq. (8), we found a value $K^c(T=100)=0.41\%$ for the Y123 chains. This result is close to the experimental value of $K^c_{\rm expt}(100)=0.334\pm 0.01\%$.[5]

However, one faces a problem in Y124. According to Fig. 1 (bottom), the experimental value of the orbital shift at $T=0$ is $K_{\rm orb}^{\rm expt}(T=0)\approx 0.24\%$, 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, $K_{\rm orb}^{c}=0.125\%$. On the other hand, $(1/T_1)_{\rm NQR}$ below T_c is only slightly affected by the onset of superconductivity [see Fig. 1 (top)].

We see two possible explanations for the disagreement between $K_{\rm orb}^{\rm expt}(0)$ and $K_{\rm orb}^{c}$. One could be that, below T_c, 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 $T=0$. The other explanation is that, 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.

In summary, based on the hypothesis of a ''Luttinger liquid'' ground state within the 1D Hubbard model, the temperature and concentration dependences of the nuclear spin-lattice relaxation and the Knight shift of the chain Cu(1) nuclei in the normal states of Y123 and Y124 compounds were calculated. 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.

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