A possible explanation of the normal-state nuclear magnetic resonance T_1^{-1} for the cuprates

1993 J. Phys.: Condens. Matter 5 9215

(http://iopscience.iop.org/0953-8984/5/49/023)

J. Phys.: Condens. Matter 5 (1993) 9215–9221. Printed in the UK

A possible explanation of the normal-state nuclear magnetic resonance T_1^{-1} for the cuprates

Mukul S Laad
Department of Physics, Indian Institute of Technology, Powai, Bombay 400076, India

Received 14 May 1993

Abstract. We seek to explain the large $T = 0$ intercept of the normal-state NMR relaxation rate of Cu nuclei in cuprate superconductors by $T = 0$ local quantum fluctuations in the $S = \frac{1}{2}$ Heisenberg model. Good quantitative agreement is obtained at both low and high T with published experimental results.

1. Introduction

The unusual magnetic properties of the quasi-2D oxide HTSCs have generated renewed interest in the spin dynamics of the $S = \frac{1}{2}$ Heisenberg antiferromagnets (HAFMs). Among other things, NMR, which is a powerful probe of the *local* spin dynamics, has demonstrated these unusual properties [1]. Generically, the oxide HTSCs show the following behaviour of $1/T_1$ for the planar Cu nuclei.

(1) $T_1^{-1} = a + bT$ over a wide range ($T^* < T < 700$ K) [2].

(2) A slight deviation from linear behaviour occurs above T_c, and the Hebel–Slichter coherence peak is absent [3].

(3) The relaxation rates are larger by a factor of more than 10 than those for the usual metals as well as those from band theory estimates.

There have been suggestions concerning the $T = 0$ intercept [4, 5]. To our knowledge, the only microscopic explanation has been that given by Baskaran and Sardar [6], who invoke the mechanism of spinon pair emission in a '2D Luttinger liquid'. The existing theories are either microscopic or phenomenological Fermi-liquid theories [7] or phenomenological marginal Fermi-liquid theories [8] and RVB theories [4].

Our aim is to show that local single-site quantum fluctuations provide an alternative mechanism for relaxing the nuclear spin at $T = 0$. Apart from [6, 8], other theories have not succeeded in providing a satisfactory explanation of a. We emphasize that it is necessary to treat local spin fluctuations adequately when one is concerned with such local probes as NMR, as the NMR T_1^{-1} is related to the transverse spin–spin correlation function at the same site [9]. Another way of looking at the problem is to note that

$$T_1^{-1} = T \lim_{\omega \to 0} \left(\sum \frac{\chi(q, \omega)}{\omega} \right). \tag{1}$$

where $\chi(q, \omega)$ is the transverse dynamical susceptibility. Hence T_1^{-1} is determined by contributions from all q up to the Brillouin zone boundary. It is easy to see why spin-wave-like theories fail to yield the constant a part of T_1^{-1}; they do not treat the large-q part

0953-8984/93/499215+07$07.50

properly. (At large q, the spin-wave damping is appreciable, and so long-lived spin waves are unstable.)

Approaches based on the Holstein–Primakoff SWT are beset with more basic problems; they are good approximations in the limit $S \gg 1$ and certainly should be used with reservations for $S = \frac{1}{2}$. However, as long as one deals with observables such as sublattice magnetization, etc, these approaches give good answers, but the inclusion of local constraints on boson occupation leads to drastically different results.

2. Calculation of the 'spin-wave density of states'

We shall focus on the local spin fluctuations; we work with the $S = \frac{1}{2}$ HAFM, with the inclusion of transverse anisotropy in the Hamiltonian for brevity. This enables us to reduce the CuO_2 plane to a 2D $S = \frac{1}{2}$ HAFM, at least as long as we concern ourselves with spin fluctuations. The basic process of nuclear relaxation arises from the usual hyperfine coupling of the nuclear and the electronic spins. It is easy to see (cf equation (1)) that a finite T_1^{-1} at $T = 0$ implies a finite density of states (DOS) for the spin excitations as the excitation energy goes to zero; this provides the channel for the nuclear spin to relax at $T = 0$.

The above is therefore closely linked to the local spin dynamics of the $S = \frac{1}{2}$ HAFM as the anisotropy is varied; we start with the $S = \frac{1}{2} XXZ$ model:

$$H = J \sum [S_i^z S_j^z + \tfrac{1}{2}\alpha(S_i^+ S_j^- + \text{HC})] \tag{2}$$

where the sum is over nearest neighbours (NNs) in this paper.

As is known, at $\alpha = 0$, the Néel states are the exact eigenstates. The XY part of the model plays an increasingly important part as α is raised; owing to spin-flip terms in equation (2), there are now many 'wrongly oriented' spins at the 'wrong' sites. We could equivalently say that the spin degrees of freedom are itinerant as α is raised. Thus, we are interested in the local dynamics of a spin situated in a background of spins fluctuating quantum-mechanically. Physically, with a low doping level, antiferromagnetic correlations are short-ranged at any non-zero T in two dimensions, with no magnetic order, and a single-site theory could be expected to provide a good description of the local dynamics. We start with the representation [10]

$$S_i^+ = b_i^+ \qquad S_i^- = b_i \qquad S_i^z = b_i^+ b_i - \tfrac{1}{2} \tag{3}$$

where the b anticommute at the same site but commute at different sites; this means that there is a hard-core constraint on the occupation of more than one b at a site. This constraint is enforced by adding a Lagrange multiplier μ, which plays the role of a chemical potential. The Hamiltonian is now written as

$$H = J \sum [n_i^b n_j^b + \tfrac{1}{2}\alpha(b_i^+ b_j + \text{HC})]. \tag{4}$$

We remark at this juncture that, as far as the single-site dynamics are concerned, we can treat the b as fermions. We use the alloy analogy approach (AAA) together with the best single-site theory, the CPA [11], to study local spin dynamics. We consider the situation where the 'b fermion' hops onto site i when the z NNs are empty, or occupied by b fermions, with respective probabilities $x = 1 - n^b$ and $y = n^b$ due to the quantum fluctuations of the background spins. The problem is reminiscent of the problem of the Anderson impurity,

i.e. a single impurity self-consistently embedded in an effective medium; self-consistency is achieved by demanding that $\langle n_i^b \rangle$ calculated self-consistently for the 'impurity' be set equal to n^b defined above.

Within the AAA, the probabilities x and y represent the alloy concentrations, while the site energies of the alloy (composed of empty sites or sites occupied by b fermions; this corresponds to sites occupied by down or up spins in the original problem) are

$$\varepsilon_1^b = -\mu \qquad \varepsilon_2^b = -\mu + zJ. \tag{5}$$

The unperturbed propagator is

$$G_0^{bb}(q, \omega) = (\omega - \varepsilon_q + \mathrm{i}\eta)^{-1}. \tag{6}$$

We choose the unperturbed DOS to be a Lorentzian with half-width Δ_b [12]. (We have checked separately for a bounded DOS and, so long as the gap is zero, our results are more or less unaffected. This occurs for $\alpha \leqslant 1$.)

The unperturbed local propagator is given by

$$G_0^{bb}(\omega) = \int \rho_0(\varepsilon) G_0^{bb}(\varepsilon, \omega)\, \mathrm{d}\varepsilon = (\omega + \mathrm{i}\Delta)^{-1}. \tag{7}$$

The perturbed propagator is calculated from Dyson's equation

$$G^{bb}(\omega) = [\omega - \Sigma^b(\omega) + \mathrm{i}\Delta_b]^{-1} \tag{8}$$

where $\Sigma^b(\omega)$ is the multiple-scattered self-energy calculated from Soven's equation [11]

$$\Sigma^b(\omega) = \varepsilon^b - [\varepsilon_1^b - \Sigma^b(\omega)] 2\pi G^{bb}(\omega) [\varepsilon_2^b - \Sigma^b(\omega)] \tag{9}$$

where $\varepsilon^b = zJn^b$. Substituting equations (5) and (8) in (9) yields the self-energy:

$$\Sigma^b(\omega) = zJn^b + (zJ)^2 n^b (1 - n^b)/[\omega - zJ(1 - n^b) + \mathrm{i}\Delta_b]. \tag{10}$$

The spin–spin Green function is now given by

$$G^{bb}(\omega) = (2\pi)^{-1}[(1 - n^b)/(\omega + \mathrm{i}\Delta_b) + n^b/(\omega - zJ + \mathrm{i}\Delta_b)]. \tag{11}$$

The DOS of the spin-wave excitations is

$$\rho^{\mathrm{SW}}(\omega) = (\Delta_b/2\pi^2)[(1 - n^b)/(\omega^2 + \Delta_b^2) + n^b/(\omega - zJ)^2 + \Delta_b^2]. \tag{12}$$

The number density is easily calculated by integrating this equation up to the chemical potential μ, and the magnetization is then easily calculated to be

$$\langle S_i^z \rangle = (1/2\pi^2) P_0[\Psi]/Q_0[\Psi] \tag{13a}$$

where

$$P_0[\Psi] = \tan^{-1}(\mu/\Delta_b) + \tan^{-1}[(\mu - zJ)/\Delta_b] \tag{13b}$$

$$Q_0[\Psi] = 1 + (1/\pi)\{\tan^{-1}(\mu/\Delta_b) - \tan^{-1}[(\mu - zJ)/\Delta_b]\}. \tag{13c}$$

We choose $\mu = \frac{1}{2}zJ$, so that $\langle S_i^z \rangle = 0$; we work with this throughout. A comment on equation (12) is in order here. The DOS is never zero in the region between the two spin-split 'bands'; this difficulty is not severe and can be taken care of by simply replacing the unperturbed DOS by a 'bell-shaped' DOS. We have done this, and we find that the spin gap closes at $\alpha = 1$ [14]. This is indicative of a transition to a 'quantum-spin-liquid'-like phase with a gapless spin excitation spectrum. This transition is smooth, and a possibility is to test this by neutron scattering. We emphasize that this is very different from a picture where spin-wave-like excitations are stable; at such short length scales, it is more probably the case that local constraints on boson occupation lead to new physics.

3. Nuclear magnetic resonance relaxation time T_1^{-1}

As mentioned earlier, the NMR T_1^{-1} shows anomalous behaviour. Baskaran and Sardar [6] have sought to provide a possible explanation for a by invoking the mechanism of spinon pair emission in a '2D Luttinger liquid', based on the hypothesis of spin-charge decoupling of the low-energy excitations. In their analysis, the two-spinon DOS is finite as the energy of the pair goes to zero; this provides a channel for the nuclear spin to relax even at $T = 0$.

We shall show that the inclusion of local quantum fluctuations provides an alternative mechanism for relaxing the nuclear spin at $T = 0$. The Hamiltonian describing the coupling of the nuclear spin to the electronic spin of the Cu is [15]

$$H = A \sum_i \boldsymbol{I}_i \cdot \boldsymbol{S}_i(0) \tag{14}$$

where $\boldsymbol{S}(0)$ is the electronic spin density at the nucleus. The value of A for the Cu on most of the CuO planes in HTSC materials is $A = 180$ kOe/μ_B. The constant a has the value 1200 s^{-1} for $YBa_2Cu_3O_{6.5}$ and 1800 s^{-1} for La_2CuO_4. The equation for the NMR relaxation time follows from the usual Fermi golden rule [16]:

$$\frac{1}{T_1} = \frac{A^2}{4\hbar^2 N} \sum_i [\langle S_i^+ S_i^- \rangle + \text{HC}] \tag{15}$$

where

$$\langle S_i^+ S_i^- \rangle = \int \langle T[S_i^+(t) S_i^-(0)] \rangle \exp(\mathrm{i}\omega t)\, \mathrm{d}t \tag{16}$$

(at $\omega = 0$). We assume that [16]

$$\langle T[S_i^+(t) S_i^-(0)] \rangle = \langle S_i^+ S_i^- \rangle_0 \exp(-t/\tau) \tag{17}$$

where $\tau = h/J$ is the characteristic time scale associated with the spin fluctuations. $\langle S_i^+ S_i^- \rangle_0$ is just the n^b calculated in section 2. Substituting equation (17) into equation (16) and the result into equation (15) yields

$$T_1^{-1} = \frac{A^2}{2\hbar J N} \sum_i \langle S_i^+ S_i^- \rangle_0. \tag{18}$$

To evaluate this, we use equation (11) for $G^{bb}(\omega)$ and perform the sum at an arbitrary temperature following Keiter [17] to obtain

$$n^b = P[\Psi]/Q[\Psi]$$

where

$$\begin{aligned} P[\Psi] &= \tfrac{1}{2} + (1/\pi)\,\mathrm{Im}\,\Psi\{1/2[1 + (\beta/\pi)(\Delta_b + \mathrm{i}\mu)]\} \\ Q[\Psi] &= 1 + (1/\pi)[[\mathrm{Im}\,\Psi\{1/2[1 + (\beta/\pi)(\Delta_b + \mathrm{i}\mu)]\} \\ &\qquad - \mathrm{Im}\,\Psi\{1/2[1 + (\beta/\pi)(\Delta_b + \mathrm{i}(\mu - zJ))]\}]] \end{aligned} \tag{19}$$

where Im Ψ is the imaginary part of the digamma function.

We look separately at the limits of high and low T.

In the low-T regime, $\beta\Delta_b \gg 1$, so that using the asymptotic form of the digamma function, we obtain

$$n^b = [\tfrac{1}{2} + (1/\pi)\tan^{-1}(\mu/\Delta_b)]/[\![1 + (1/\pi)\{\tan^{-1}(\mu/\Delta_b) - \tan^{-1}[(\mu - zJ)/\Delta_b]\}]\!] \tag{20}$$

and hence, as $T \to 0$, the inverse relaxation time is given by

$$T_1^{-1} = A^2 n^b / 2\hbar J. \tag{21}$$

We work with a singlet ground state, so that $n^b = \frac{1}{2}$, and from equation (20) we have $\mu = \frac{1}{2}zJ$. We set $J = 0.15$ eV and consider the isotropic limit, so that $\alpha = 1$. Putting $A = 180$ kOe/μ_B, we calculate $T_1^{-1} = 1316$ s^{-1}. The experimental value is 1200 s^{-1}. The agreement is good, considering that anisotropy and geometrical factors are completely neglected, and that our form of the hyperfine interaction is rather approximate.

In the high-T regime, $\beta\Delta_b \to 0$, so that using the asymptotic form of the digamma function in this regime, we obtain

$$T_1^{-1} = (A^2/2\hbar J)[\tfrac{1}{2} + \tfrac{1}{2}\tanh(\tfrac{1}{2}\beta\mu)]/[\![1 + \tfrac{1}{2}\{\tanh(\tfrac{1}{2}\beta\mu) - \tanh[\tfrac{1}{2}\beta(\mu - zJ)]\}]\!] \tag{22}$$

and so, in the regime $\beta\mu \to 0$, $\beta zJ \to 0$, equation (22) reduces to

$$T_1^{-1} = (A^2/2\hbar J)(\mu + 2k_B T)/(zJ + 4k_B T) \tag{23}$$

consistent with the $T/(\alpha + \beta T)$ form observed by Kitaoka *et al* [18]. Thus, we observe good agreement with published experimental data in both the low- and the high-T regimes.

4. Discussion

Our aim has been to explain the extrapolated $T = 0$ contribution to T_1^{-1}. Earlier calculations with the exception of [6, 8] do not seem to yield a satisfactory explanation of this part. We have suggested that an adequate treatment of local fluctuations by employing the best single-site theory, the CPA, provides a possible alternative explanation. We have succeeded in obtaining very good agreement in both high- and low-T limits with published experimental results. Our analysis here basically describes the following picture. The nuclear spin relaxes via the usual hyperfine coupling to the electronic spin. The fact that we have a non-zero DOS for the spin excitations as $\omega \to 0$ provides a channel for the nuclear spin to relax even at $T = 0$, leading to the observed $T = 0$ contribution. The Korringa mechanism would, on the other hand, arise from the spin-flip scattering of thermally excited spin excitations, yielding a zero contribution at $T = 0$. This also yields the linear-in-T 'Korringa' behaviour at finite T. Our results remain qualitatively unaffected as long as the DOS remains gapless. We have checked this for the case of a bounded unperturbed DOS by replacing the Lorentzian by a bell-shaped DOS:

$$\rho_0(\omega) = \begin{cases} (2/\pi\Delta^2)(\Delta^2 - \omega^2)^{1/2} & \text{for } -\Delta < \omega < \Delta \\ 0 & \text{otherwise.} \end{cases} \tag{24}$$

The 'spin gap' now closes at $\alpha = \alpha_c < 1$ so that, in the isotropic limit, $\rho^b(0)$ is finite, and so our results are essentially unaffected. However, for $\alpha < \alpha_c$, a spin gap opens up at

$\omega = 0$ and, for such cases, we expect a change in the T-dependence of the relaxation time. Our analysis also shows that the large-q contribution is appreciable; indeed, if we calculate

$$n^b(q) = \int [\omega - \varepsilon(q) - \Sigma^b(\omega)]^{-1}\, d\omega \tag{25}$$

we see that it has considerable large-q contributions, where long-lived spin excitations are no longer stable elementary excitations. Hence, if for instance we were to start from Holstein–Primakoff SWT, we would have to retain higher-order terms in $a^+a/2S$, to introduce coupling between various q modes and to enforce the constraint on boson occupation for $S = \frac{1}{2}$. Our treatment avoids the first difficulty, but we end up with the problem of treating the spins as fermions at the same site, and as bosons on different sites. However, as far as single-site correlators are concerned, we can still treat the spins as fermions, which resolves the difficulty.

Our analysis would be closest to reality in lightly doped antiferromagnets, where long-range order has more or less given way to a disordered antiferromagnetic state with a short antiferromagnetic correlation length of order 10–15 Å. Recent experimental work [19] has shown that, in lightly doped 2:1:4 compounds, the T-dependence of $\chi(q, \omega)$ is solely due to the T-dependence of the local spin response. This does not contradict, but rather supports, our view that a good treatment of the local response is required for a proper description of magnetic fluctuations in these materials. This lends good support to the point of view that we have adopted in this paper. The part (2) in the introduction requires a more careful investigation; it is observed that, at $T_c < T < T^*$, a characteristic scale slightly above T_c, T_1^{-1} shows a deviation from linearity, and just above T_c there is no coherence peak that one would associate with BCS superconductors. This is most probably indicative of a change in the local spin response just above T_c, but more work has to be done to check this point. Related work is in progress.

Acknowledgments

I would like to thank Professor G Baskaran and Manas Sardar for providing a copy of their paper prior to publication. I would also like to thank Professor D K Ghosh for reading the manuscript, and for constructive criticism.

References

[1] Kitaoka Y *et al* 1989 *Phys. Rev. Lett.* **63** 1865
[2] Kitaoka Y, Ishida K, Ohsugi S, Fujiwara K and Asayama K 1991 *Physica* C **185–9** 98
Hemmel P C *et al* 1989 *Phys. Rev. Lett.* **63** 1288
Alloul H, Ohno T and Mendels P 1989 *Phys. Rev. Lett.* **63** 1700
Imai T *et al* 1989 *Physica* C **162–4** 169
[3] Warren W W, Walstedt R E, Brennert G F, Bell R F, Espinosa G P and Cava R J 1989 *Physica* C **162–4** 179
[4] Anderson P W 1989 *Phys. Rep.* **184** 2–4
[5] Bulut N, Hone D, Scalapino D J and Bickers N E 1990 *Phys. Rev.* B **41** 1797
Moriya T 1990 *J. Phys. Soc. Japan* **59** 2905
[6] Baskaran G and Sardar M 1993 *IMSc Preprint*; 1993 *Phys. Rev. Lett.* submitted
Fukuyama H and Kuboki K 1990 *J. Phys. Soc. Japan* **59** 2617
[7] Pines D 1992 *Physica* C **185–9** 120
Millis A J, Monien H and Pines D 1990 *Phys. Rev.* B **42** 167

[8] Varma C M, Littlewood P, Schmitt-Rink S, Ruchenstein A and Abrahams E 1989 *Phys. Rev. Lett.* **69** 1996
[9] White R M *Magnetism* vol 32 (Berlin: Springer)
[10] Zubarev D N 1960 *Usp. Fiz. Nauk.* **71** 71
[11] Economou G *Green Functions in Quantum Physics* vol 7 (Berlin: Springer)
Velicky B, Kirkpatrick S and Ehrenreich H 1968 *Phys. Rev.* **175** 747–68
[12] Georges A and Kotliar G 1991 *Proc. Adriatico Research Workshop on Physics of Strongly Correlated Electronic Systems* (Trieste: International Center of Theoretical Physics)
[13] Economou G *Green Functions in Quantum Physics* vol 7 (Berlin: Springer)
[14] Laad M S 1993 unpublished
[15] Shastry B S 1988 *Phys. Rev. Lett.* **63** 1288
Mila F and Rice T M 1989 *Physica* C **157** 561
[16] White R M *Magnetism* vol 32 (Berlin: Springer)
[17] Keiter H 1970 *Phys. Rev.* B **2** 3777–91
[18] Kitaoka Y *et al* 1989 *Phys. Rev. Lett.* **63** 1865
[19] Hayden S M *et al* 1991 *Phys. Rev. Lett.* **66** 821–5