Theory for the Gaussian Component of the ^{63}Cu Nuclear Spin-Echo Decay Rate $1/T_{2G}$ in $La_{2-x}Sr_xCuO_4$

I. A. Larionov [1,2,*]

Email Larionov.MRSLab@mail.ru

[1] National University of Science and Technology “MISIS”, Leninskiy pr. 4, 119049 Moscow, Russia

[2] Institute of Physics, Kazan Federal University, Kremlevskaya Str. 18, 420008 Kazan, Russia

Abstract

We report calculations of the Gaussian component of the ^{63}Cu nuclear spin-echo decay rate $1/T_{2G}$ employing the theory for spin susceptibility as derived within the *t-J* model starting from carrier-free La_2CuO_4 and right up to optimally doped superconducting layered copper oxide $La_{2-x}Sr_xCuO_4$. The theory reproduces the temperature and doping behavior of the experimental data for $1/T_{2G}$. A quantitative agreement with experimental $1/T_{2G}$ data in doped $La_{2-x}Sr_xCuO_4$ compounds is obtained with account of both the spin–spin and “fermion”–“fermion” correlations.

1. Introduction

It has been firmly established soon after the discovery of superconductivity in layered copper oxides that the parent, carrier-free compounds, are the two dimensional $S = 1/2$ Heisenberg antiferromagnetic (AF) insulators. The superconducting properties appear with doping, concentration of charge carriers per plane copper site, $\delta > 0.04$, reaching a maximum T_c at optimal doping $\delta_{opt} \approx 0.15$. The magnetic properties also undergo the dramatic changes: the AF long

range order in the carrier-free compound is lost upon doping $\delta > 0.02$, however, the AF short range order is still present and decreases with doping and temperature. In this paper we will discuss the evolution of AF order with doping and temperature and approve the expression for AF correlation length by comparison with the Gaussian component of the ^{63}Cu nuclear spin-echo decay rate $1/T_{2G}$ data. The measurements of the transverse relaxation rate $1/T_2$ in high temperature superconductors (high-T_c) provide important information concerning the static spin susceptibility $\chi_{\mathbf{k}}$, which is complementary to the information obtained from nuclear spin–lattice relaxation rate $1/T_1$ [1–5].

2. Basic relations

The Gaussian component of the ^{63}Cu nuclear spin-echo decay rate $1/T_{2G}$ in nuclear quadrupole resonance (NQR) experiments is given by [5],

$$^{63}T_{2G}^{-2} = \frac{0.691}{64\hbar^2\mu_B^4}\left\{\sum_k F_\perp(k)^2\chi_k^2 - [\sum_k F_\perp(k)\chi_k]^2\right\}, \quad (1)$$

where $F_\perp(\mathbf{k}) = [A_c + 4B\gamma_{\mathbf{k}}]^2$ is the hyperfine form factor, with $A_c = -17.25 \times 10^{-7}$ eV being the direct, on-site coupling of the ^{63}Cu nuclei to the Cu^{2+} spins, and $B = (1 + 2.75\delta) \times 3.37 \times 10^{-7}$ eV is the strength of the transferred hyperfine coupling of the Cu^{2+} nuclear spin to the four nearest neighbor (NN) copper spins [3, 4]. The factor 0.691 comes from the natural abundance of ^{63}Cu nucleus, $\gamma_{\mathbf{k}} = (1/2)(\cos k_x + \cos k_y)$, and μ_B is the Bohr magneton. Note that Eq. (1) assumes that the nuclear spins over the entire spectrum are flipped by the π pulse, a condition that is not satisfied in $La_{2-x}Sr_xCuO_4$ [5]. Since A_c and B constants do not depend on T, the temperature dependence of $1/T_{2G}$ is fully described by the temperature dependence of static spin susceptibility, $\chi_{\mathbf{k}}$.

The analytical expression for static spin susceptibility has similar structure at any doping level [6–8] as in the isotropic spin-wave theory [9]. Our interest to it is caused by the fact that the structure of analytical expression for static spin susceptibility remains nearly

unchanged in numerous theories for spin susceptibility with the minor difference as the additional term in the numerator only.

We will consider two very close expressions for static spin susceptibility. The first one has been independently obtained in [6, 7]

$$\chi_k = \frac{4|c_1|}{Jg_-(g_+ + \gamma_k)}. \qquad 2$$

The parameter g_+ is related to antiferromagnetic correlation length ξ via the expression

$$\xi = \frac{1}{2\sqrt{g_+ - 1}}, \qquad 3$$

$J = 0.12$ eV is the AF coupling constant between the NN, the NN spin–spin correlation function is given by $c_1 = \frac{1}{4}\sum_\rho \langle S_i^z S_{i+\rho}^z \rangle$, and the index ρ runs over NN. The values of the parameters of the theory: c_1 and g_- are given in Table 1.

Table 1

The calculated in the $T \to 0$ limit antiferromagnetic spin–spin correlation function between the nearest neighbors c_1, the parameter g_-, and the spin stiffness constant ρ_S

Doping	c_1	g_-	$2\pi\rho_s/J$
$\delta = 0$	−0.1152	4.4148	0.38
$\delta = 0.12$	−0.0758	3.252	0.20
$\delta = 0.15$	−0.0617	2.947	0.13

On the other hand the static spin susceptibility χ_k is related to a generalized mean-field spin excitation spectrum ω_k via the equation

$$\chi_k = \langle [i\dot{S}_k^+, S_{-k}^-] \rangle / \omega_k^2 \qquad 4$$

where

within the model, and T_1 is the thermodynamic correlation function given by

$$T_1 = p\sum_k \gamma_k f^h_{E_k}.$$

The hopping integral, $t = J/0.3$, between NN describes the motion of electrons causing a change in their spins and

$$f^h_{E_k} = \frac{1}{\exp(-E_k + \mu)/k_B T + 1}$$

is the Fermi function of holes. The excitation spectrum of holes is given by, $E_{\mathbf{k}} = 4t_{\text{eff}}\gamma_{\mathbf{k}}$, where the hoppings, t, are affected by electronic and AF spin–spin correlations c_1, resulting in *effective* values, for which we set $t_{\text{eff}} = \delta J/0.3$ [8–11], to match the insulator–metal transition. The chemical potential μ is related to δ by $\delta = p\sum_k f^h_{E_k}$, where $p = (1 + \delta)/2$.

This result implies that

$$\chi_k = \frac{4J|c_1| + 2t|T_1|}{J^2 g_-(g_+ + \gamma_k)}, \qquad 5$$

which differs from Eq. (2) by the additional term $2t|T_1|$ in the numerator only.

According to [6], the temperature and concentration dependence of the correlation length ξ is given by

$$\xi = \frac{J\sqrt{g_-}}{\theta_1}[1 - \exp(-\theta_1/k_B T)]\exp(2\pi\rho_s/k_B T), \qquad 6$$

where ρ_S is spin stiffness, whose calculated value is given in the Table 1,

7

$$\theta_1^2 = \frac{32t_{\mathrm{eff}}^2(1+\delta)}{\pi}\int_{-1}^{1} d\gamma \frac{\gamma^2 K(\sqrt{1-\gamma^2})f_\gamma^h}{\gamma^2+\tilde{\omega}_1^2} \times \frac{\tilde{\omega}_1\sqrt{1+\tilde{\omega}_1^2}}{K(1/\sqrt{1+\tilde{\omega}_1^2})} - 2Jv_1,$$

$$v_1 = \frac{8t_{\mathrm{eff}}(1+\delta)}{\pi}\int_{-1}^{1} d\gamma \frac{\gamma K(\sqrt{1-\gamma^2})f_\gamma^h}{\gamma^2+\tilde{\omega}_1^2} \times \frac{\tilde{\omega}_1\sqrt{1+\tilde{\omega}_1^2}}{K(1/\sqrt{1+\tilde{\omega}_1^2})}, \quad (8)$$

where δ is the number of *extra* holes, due to doping, per one plane Cu^{2+}, which can be identified with the Sr content x in $La_{2-x}Sr_xCuO_4$, $K(x)$ is a complete elliptic integral, $\tilde{\omega}_1 = \omega_1/4t(1+\delta)$, and $\omega_1 = 2\pi k_B T$.

For small δ, one finds $\theta_1 \sim \sqrt{\delta T}$ and the expression (4) smoothly approaches the result for the Heisenberg antiferromagnet, i.e.:

$$\xi \sim \frac{J\sqrt{g}}{k_B T}\exp(2\pi\rho_s/k_B T). \quad (9)$$

One should note that in the carrier-free regime the derivation of ξ has been performed from exponential decay of the spin–spin correlation function at large separations, and at finite doping it can be derived from expansion of the static spin susceptibility around the AF wave vector $\mathbf{Q} = (\pi, \pi)$, yielding the same expression as that given by Eq. (6) [6,12].

Before plotting $1/T_{2G}$ as a function of T we should pay attention to the fact that according to Eq. (6) the correlation length diverges at $T = 0$, even in doped compounds, which is in contrast with neutron scattering data [16, 17]. This disagreement is supposed to be connected with the Kondo–Yamaji [18] decoupling procedure used for the derivation of the susceptibility, which, probably, overestimates the role of AF correlations at low temperatures. Thus, to avoid the divergence we replace ξ by ξ_{eff}, which is given by:

$$\xi_{\mathrm{eff}}^{-1} = \xi_0^{-1} + \xi^{-1}, \quad (10)$$

where a fair agreement with neutron scattering (NS) data [16, 17] around the optimal doping is obtained with $\xi_0 = 1/\delta$ [19]. Having established the temperature and doping dependence of the correlation

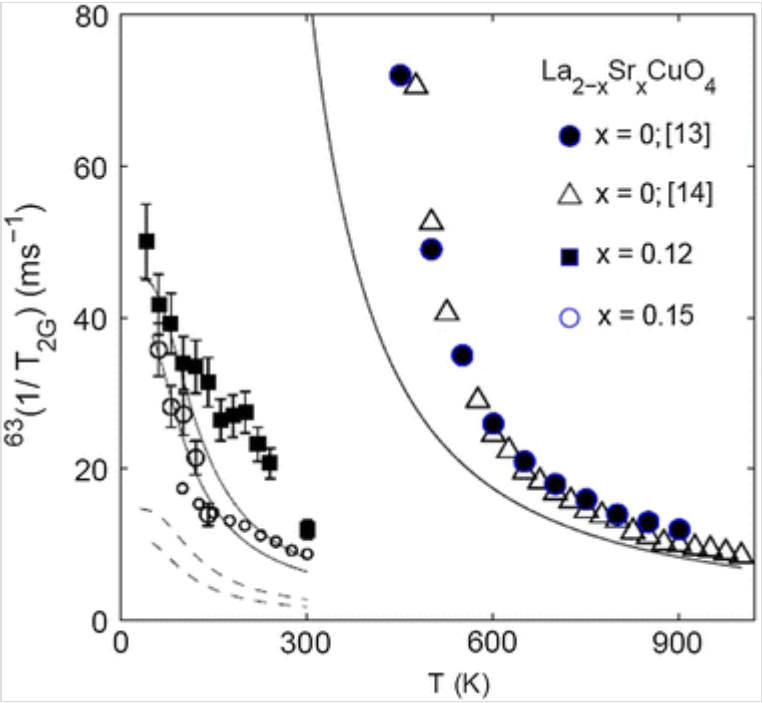
length we now proceed with calculations of the Gaussian component of the ^{63}Cu nuclear spin-echo decay rate $1/T_{2G}$.

3. Comparison with experiment and discussion

Figure 1 shows the calculated temperature and doping dependencies of the Gaussian component of the ^{63}Cu nuclear spin-echo decay rate $1/T_{2G}$ without any adjustable parameters. We found that $1/T_{2G}$ increases monotonously with decreasing temperature in agreement with experimental data [5, 13–15]. It is seen that the $1/T_{2G}$ temperature dependence is similar in both carrier-free and doped $La_{2-x}Sr_xCuO_4$, opposite to plane copper $1/T_1$, which shows a wide peak at temperatures around 150 K and at moderate doping.

Fig. 1

Temperature dependence of the Gaussian component of the ^{63}Cu nuclear spin-echo decay rate $1/T_{2G}$ for carrier-free La_2CuO_4 from Ref. [13] (*filled circles*) and Ref. [14] (*open triangles*), and for doped $La_{2-x}Sr_xCuO_4$ (*filled squares* for $x = 0.12$ and *open circles* for $x = 0.15$) from Ref. [5]. *Small open circles* without *error bars* show the results of Ref. [15] for $x = 0.15$. *Lower* and *upper solid lines* show results of the calculations for $x = 0.12$ and for $x = 0.15$, respectively, with the static spin susceptibility given by Eq. (5) that accounts for the "fermionic" correlation function factor. *Lower* and *upper dashed lines* show the results of the calculations for $x = 0.12$ and for $x = 0.15$, respectively, without the "fermionic" correlation function factor in the numerator of the expression for static spin susceptibility (Eq. 2)



In high-T_c cuprates the AF correlations are very strong and this leads to necessity of some scaling relation analysis that relates the temperature dependencies of magnetic quantities with that of AF correlation length and the characteristic energy of the AF spin fluctuations ω_{sf}. Both these quantities are related through the dynamical exponent z as $\omega_{sf} \propto \xi^{-z}$. From both Eqs. (2) and (5) it follows that $\chi(Q = (\pi, \pi)) \propto \xi^2$. Most experiments on high-T_c materials show $z = 1$ [5, 13, 15, 20]. At the quantum critical point $z = 1$ is expected while $z = 2$ is believed for AF spin fluctuations in itinerant electron systems. However, Keren et al. [21] showed that dynamical fluctuations of copper nuclei including both spin–lattice and flip-flop processes in the analysis of nuclear magnetic resonance (NMR) transverse relaxation data gives $z = 1$. Both AF $x = 0$ and doped cases showed that $1/T_{2G}$ data are consistent with $z = 1$.

A similar increase of $1/T_{2G}$ with decreasing temperature above T_c is observed in $YBa_2Cu_4O_8$ [22] with nearly the same absolute values considering that the doping level in $YBa_2Cu_4O_8$ is below optimal ($x = 0.15$) and is around $x = 0.12$.

AQ1

4. Conclusion

In conclusion, we have calculated the temperature and doping dependence of the Gaussian component of the ^{63}Cu nuclear spin-echo decay rate $1/T_{2G}$ taking into account the temperature and doping behavior of the AF correlation length. We obtain a fair agreement with the experimental data in the carrier-free and doped cases without adjustable parameters in a wide temperature and doping range. Both theoretical and experimental research is required to elucidate the meaning of the dynamical exponent z in copper oxide superconductors. Both the spin–spin and "fermion"–"fermion" correlations shall be used to explain quantitatively the experimental $1/T_{2G}$ data in doped $La_{2-x}Sr_xCuO_4$ compounds.

Acknowledgments

The work was carried out with financial support from the Ministry of Education and Science of the Russian Federation in the framework of Increase Competitiveness Program of NUST "MISiS'' (No. K2-2014-015).

References

1. D. Thelen, D. Pines, Phys. Rev. B **49**, 3528 (1994)

2. M. Takigawa, Phys. Rev. B **49**, 4158 (1994)

3. A.J. Millis, H. Monien, D. Pines, Phys. Rev. B **42**, 167 (1990)

4. Y. Zha, V. Barzykin, D. Pines, Phys. Rev. B **54**, 7561 (1996)

5. S. Fujiyama, M. Takigawa, Y. Ueda, T. Suzuki, N. Yamada, Phys. Rev. B **60**, 9801 (1999)

6. AYu. Zavidonov, D. Brinkmann, Phys. Rev. B **58**, 12486 (1998)

7. M.V. Eremin, A. Aleev, I. Eremin, Zh Eksp, Teor. Fiz. **133**, 862 (2008);

8. [M.V. Eremin, A. Aleev, I. Eremin, JETP **106**, 752 (2008). (Engl. Transl.)]

9. A. Sokol, R.R.P. Singh, N. Elstner, Phys. Rev. Lett. **76**, 4416 (1996)

10. P.W. Anderson, Science **235**, 1196 (1987)

11. G. Baskaran, Z. Zou, P.W. Anderson, Solid State Commun. **63**, 973 (1987)

12. I.A. Larionov, Phys. Rev. B **69**, 214525 (2004)
AQ2

13. M.E. Fisher, Rep. Prog. Phys. **30**, 615 (1967)

14. T. Imai, C.P. Slichter, K. Yoshimura, M. Kato, K. Kosuge, Phys. Rev. Lett. **71**, 1254 (1993)

15. M. Matsumura, H. Yasuoka, Y. Ueda, H. Yamagata, Y. Itoh, J. Phys. Soc. Jpn. **63**, 4331 (1994)

16. R.E. Walstedt, S.-W. Cheong, Phys. Rev. B **53**, 6030 (1996)

17. M.A. Kastner, R.J. Birgeneau, G. Shirane, Y. Endoh, Rev. Mod. Phys. **70**, 897 (1998)

18. G. Aeppli, T.E. Mason, S.M. Hayden, H.A. Mook, J. Kulda, Science **278**, 1432 (1997)

19. J. Kondo, K. Yamaji, Prog. Theor. Phys. **47**, 807 (1972)

20. I.A. Larionov, Phys. Rev. B **72**, 094505 (2005)

21. R.L. Corey, N.J. Curro, K. O'Hara, T. Imai, C.P. Slichter, K. Yoshimura, M. Katoh, K. Kosuge, Phys. Rev. B **53**, 5907 (1996)

22. A. Keren, H. Alloul, P. Mendels, Y. Yoshinari, Phys. Rev. Lett. **78**, 3547 (1997)

23. A. Sokol, R.L. Glenister, R.R.P. Singh, Phys. Rev. Lett. **72**, 1549 (1994)