Relaxation function theory for spin dynamics of strongly correlated layered copper oxide superconductors

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Abstract. We present the relaxation function theory for dynamic spin susceptibility for doped two-dimensional \( S = \frac{1}{2} \) Heisenberg antiferromagnetic (AF) system in the paramagnetic state as obtained by means of the Mori-Zwanzig projection operator procedure to the \( t-J \) model. The results of the calculations are discussed in connection with the peculiar properties of layered copper oxide high-temperature superconductors (high-\( T_c \)). These include the neutron resonance peak, pseudogap properties, \( \omega/T \) scaling, and the temperature and doping dependence of plane copper and oxygen nuclear spin-lattice relaxation rates (NSLRR). Particularly, the role of AF short range order, its evolution with doping and saturation of AF correlation length at low temperatures, is highlighted in view of the dynamic spin response of high-\( T_c \) up to optimal (maximum \( T_c \)) doping. The contribution from spin diffusion to relaxation rates is evaluated and is shown to play a dominant role in plane oxygen NSLRR of lightly doped (\( \sim 3\% \) holes per copper site) high-\( T_c \) at low temperatures. It is shown that the spin-wave-like theory is able to reproduce the main features of spin dynamics in high-\( T_c \) as observed experimentally.

1. Introduction
Dynamic phenomena in strongly correlated electron systems (SCES) continues to be in the focus of attention by condensed matter physicists due to rich variety of properties ranging from high temperature superconductivity (high-\( T_c \)) in layered copper oxides and exotic phases in other low-dimensional materials [1, 2, 3, 4, 5, 6]. Despite of considerable both the experimental and theoretical effort these systems still hide a host of undisclosed properties. Theoretical studies of dynamical properties [7, 8, 9, 10] make use of auxiliary quantities, viz. the Green’s and relaxation function methods which have been brilliantly and exhaustively reviewed by Balucani, Lee, and Tognetti [11] in connection with both the quantum (e.g., described by Heisenberg Hamiltonian) and classical many-body systems.

Here we will discuss the spin dynamics of doped by holes a two dimensional \( S=1/2 \) Heisenberg antiferromagnetic (2DHAF) system in the paramagnetic state and compare the theoretical results obtained by Mori-Zwanzig projection operator procedure with magnetic resonance and inelastic neutron scattering data in layered copper high-\( T_c \). The relation between the Kubo’s relaxation function and the Green’s function is discussed in [11].

Inelastic neutron scattering (NS) is a powerful tool in the high frequency studies of wave vector \( k \) and frequency \( \omega \) dependent dynamic spin susceptibility \( \chi''(k, \omega) \). Emergent observations of
the resonance peak [12] at AF wave vector $Q = (\pi, \pi)$ and frequency $\omega_c \approx 40$ meV in optimally doped YBa$_2$Cu$_3$O$_{7-\delta}$, and the $\omega/T$ scaling of the averaged over the Brillouin zone the imaginary part of dynamic spin susceptibility, $\chi''(\omega, T) = \int \chi''(\epsilon, \omega, T) d^2q \approx \chi''(\omega, T \to 0)f(\omega/T)$, in the underdoped high-$T_c$ compounds [2, 13] are awaiting for theoretical understanding.

Soon after the discovery of high-$T_c$ in La$_{2-x}$(Ba,Sr)$_x$CuO$_4$ family compounds it has been established that the parent, carrier free La$_2$CuO$_4$ compound is the 2DHAL insulator in which the magnetic correlation length $\xi$ is described, e.g., by a quantum nonlinear $\sigma$ model [14, 15] and by the isotropic spin-wave theory [16] in accord with NS experiments [17]. The majority of theories for resonance peak predominatingly treat it as a feedback of superconductivity (SC) which arises in the d-wave channel [18, 19, 20, 21, 22, 23]. However, the spin-wave like features of spin susceptibility in the underdoped YBa$_2$Cu$_3$O$_{6+x}$ above $T_c$ have been emphasized [24, 25, 26, 27], suggesting that the resonant features may be caused not only by the emergence of the SC state. This conclusion has been encouraged by the absence of the isotope effect in the resonance peak frequency [28], contrary to the prediction within the RPA approach with d-wave SC [22]. In addition, Hwang, Timusk, and Gu [29] have recently shown by means of infrared spectroscopy that the resonance peak disappears completely in the overdoped Bi$_2$Sr$_2$CaCu$_2$O$_{8+y}$ sample with $T_c = 55$ K, thus suggesting the magnetic origin of the resonance peak. Thus the spin-wave concept appears to provide a natural elementary excitation in doped high-$T_c$ cuprates:

In this report, using the Mori-Zwanzig projection operator procedure with a three pole approximation for the relaxation function [11, 37, 38, 39], we will show that the resonant features in doped 2DHAL, as well as the nearly universal $\omega/T$ dependence of $\chi''(\omega, T)$ may be explained within a spin-wave-like theory [40, 41], where the correlation length, its doping dependence and saturation with lowering temperature, governs the main features of $\chi''(k, \omega)$ as observed experimentally and no SC will be presupposed in the present calculations.

2. Basic relations

2.1. The $t$–$J$ model Hamiltonian

We employ the $t$ – $J$ Hamiltonian[42] since it is the minimal model for the electronic properties of high-$T_c$ cuprates:

$$H_{t-J} = \sum_{i,j,\sigma} t_{ij} X_i^{\sigma 0} X_j^{0\sigma} + J \sum_{i>j} (S_i S_j - \frac{1}{4} n_i n_j),$$

(1)

written in terms of the Hubbard operators $X_i^{\sigma 0}$ that create an electron with spin $\sigma$ at site $i$ and $S_i$ are spin-1/2 operators. Here, the hopping integral $t_{ij} = t$ between the nearest neighbors (NN) describes the motion of electrons causing a change in their spins and $J = 0.12$ eV is the NN AF coupling constant. The spin and density operators are defined as follows:

$$S_i^\sigma = X_i^{\sigma \sigma'}, \quad S_i^z = \frac{1}{2} \sum_\sigma \sigma X_i^{\sigma \sigma}, \quad n_i = \sum_\sigma X_i^{\sigma \sigma}, \quad (\sigma = -\sigma'),$$

(2)

with the standard normalization $X_i^{\sigma \sigma} + X_i^{\sigma' \sigma'} + X_i^{\sigma' -\sigma'} = 1$. 




We formulate our study of the spin fluctuations following Mori [37], who showed its efficiency for the relaxation function.

2.2. Mori’s projection operator procedure and a three pole approximation for the dynamic relaxation function

We formulate our study of the spin fluctuations following Mori [37], who showed it’s efficiency for both the classical (and essential equivalence to Brownian motion) and quantum (e.g. Heisenberg systems of arbitrary dimension) many body systems [11]. The time evolution of a dynamical variable \( S_k^z(\tau) \), say, is given by

\[
\dot{S}_k^z(\tau) = \frac{dS_k^z(\tau)}{d\tau} = iLS_k^z(\tau).
\]  

(3)

The Liouville superoperator \( L \) represents the Poisson bracket in the classical case and in the quantal case, which is the case of the present study, it corresponds to the commutator with the Hamiltonian (1): \( iLS_k^z(\tau) \rightarrow [H_{\tau-\eta}, S_k^z(\tau)] \). The projection of the vector \( S_k^z(\tau) \) onto the \( S_k^z(\tau = 0) \) axis is given by:

\[
P_0S_k^z(\tau) = R(k, \tau) \cdot S_k^z,
\]  

(4)

and defines the linear projection Hermitian operator \( P_0 \). One may separate \( S_k^z(\tau) \) into the projective and vertical components with respect to the \( S_k^z \) axis:

\[
S_k^z(\tau) = R(k, \tau) \cdot S_k^z + (1 - P_0)S_k^z(\tau),
\]  

(5)

where

\[
R(k, \tau) \equiv (S_k^z(\tau), (S_{-k}^z)^*) \cdot (S_k^z, (S_{-k}^z)^*)^{-1}
\]  

(6)

is the relaxation function in the inner-product bracket notation:

\[
(S_k^z(\tau), (S_{-k}^z)^*) \equiv k_BT \int_0^{1/k_BT} d\phi (e^{\phi H} S_k^z(\tau) e^{-\phi H} (S_{-k}^z)^*),
\]  

(7)

and the angular brackets denote the thermal average.

For future evaluations, it is convenient to introduce a set of quantities \( f_0(\tau), f_1(\tau), \ldots, f_j(\tau), \ldots \) defined by equations: \( f_j(\tau) \equiv \exp(iL_j\tau)f_j \equiv \exp(iL_j\tau)iL_jf_{j-1} \), where \( L_0 \equiv L, f_0(\tau) \equiv S_k^z(\tau), L_j \equiv (1 - P_{j-1})L_{j-1}, \) and \( \Delta^2 \equiv (f_j, f_j^*) \cdot (f_{j-1}, f_{j-1}^*)^{-1} \) for \( j \geq 1 \). The set \( \{f_j\} \) forms an orthogonal set. The larger number of \( f_j \) is used, the finer description of \( S_k^z(\tau) \) is obtained. The last quantity from this set \( f_n \), affected by evolution operator \( \exp(iL_n\tau) \), resulting in \( f_n(\tau) \),

\[\text{Figure 1. Schematic relation between random forces } f_1(\tau) \text{ and } f_2(\tau), \text{ and their effect on the fluctuating variable } f_0(\tau) \equiv S_k^z(\tau).\]
is called the "n-th order random force" [37] acting on the variable $S_z^\omega(k)$ and is responsible for fluctuation from its average motion. 

In terms of Laplace transform of the relaxation function, $R(k, \tau)$, one may construct a continued fraction representation for $R^L(k, s)$, for which Lovesey and Meserve [39, 11] suggested a three pole approximation, 

$$R^L(k, s) = \int_0^\infty d\tau \ e^{-s\tau} R(k, \tau) \approx 1/[s + \Delta^2_{1k}/(s + \Delta^2_{2k}/(s + 1/\tau_k))], \quad (8)$$

with a cutoff characteristic time $\tau_k = 2/(\pi \Delta^2_{2k})$, by arguing that $S_z^\omega(k)$ fluctuations are weakly affected by the higher order random forces. For the relaxation shape function $F(k, \omega) = Re[R^L(k, i\omega)]/\pi$, this gives 

$$F(k, \omega) = 1/[\omega \tau_k (\omega^2 - \Delta^2_{1k} - \Delta^2_{2k})^2 + (\omega^2 - \Delta^2_{1k})^2], \quad (9)$$

where $\Delta^2_{1k}$ and $\Delta^2_{2k}$ are related to the frequency moments 

$$\langle \omega^2_k \rangle = \int_{-\infty}^{\infty} d\omega \ \omega^n F(k, \omega) = \frac{1}{\pi^n} \left[ \frac{d^n R(k, \tau)}{d\tau^n} \right]_{\tau=0}, \quad (10)$$

of $R(k, \tau)$ as $\Delta^2_{1k} = \langle \omega^2_k \rangle$, $\Delta^2_{2k} = (\langle \omega^4_k \rangle / \langle \omega^2_k \rangle) - \langle \omega^2_k \rangle$ for $\tau > \tau_k$. Note that $F(k, \omega)$ is real, even in both $k$ and $\omega$, and normalized to unity $\int_{-\infty}^{\infty} d\omega F(k, \omega) = 1$.

Here we will discuss two approximations for the imaginary part of the dynamic spin susceptibility $\chi''(k, \omega)$. The first one is in the undamped spin-wave approximation, where $F(k, \omega)$ is related to $\chi''(k, \omega)$ as 

$$\chi''_F(k, \omega) = \omega \chi(k) F(k, \omega). \quad (11)$$

Within the second one, since the relaxation function can be understood within the spin-wave framework,[11] the temperature and doping dependence of the damping of the spin-wavelike excitations may be studied further. The spin-wavelike dispersion, renormalized by interactions, is given by the relaxation function,[11] 

$$\omega^sw = 2 \int_{0}^{\infty} d\omega \ \omega F(k, \omega), \quad (12)$$

where the integration over $\omega$ in Eq. (12) has been performed analytically and exactly. One may assume the Lorentzian form for the imaginary part of the dynamic spin susceptibility, 

$$\chi''_L(k, \omega) = \frac{\chi(k) \omega \Gamma_k}{[\omega - \omega^sw]^2 + \Gamma^2_k} + \frac{\chi(k) \omega \Gamma_k}{[\omega + \omega^sw]^2 + \Gamma^2_k}, \quad (13)$$

for $k$ around the AF wave vector $(\pi, \pi)$, where the wave vector dependence of the damping is given by $\Gamma = \sqrt{\langle \omega^2_k \rangle - \langle \omega^sw \rangle^2}$.

The expression for the second moment is straightforward, 

$$\langle \omega^2_k \rangle = i \langle [\hat{S}_k^z, S^\omega_k] \rangle / \chi(k) = - (8Jc_1 - 4t_{eff}T_1) (1 - \gamma_k) / \chi(k), \quad (14)$$

where $\gamma_k = \frac{1}{z} \sum_{\rho} \exp(i k \rho) = \frac{1}{z} (\cos k_x a + \cos k_y a)$, where $a=3.8$ Ås a lattice unit and $z = 4$ is the number of nearest neighbours on the square lattice, while it is rather cumbersome for the fourth moment $\langle \omega^4_k \rangle = i \langle [\hat{S}_k^z, S^\omega_k] \rangle / \chi(k)$ and has been calculated with the approximations using the
decoupling procedures for thermal averages as will be described in following Subsection. The final result is

\[
\langle \omega_k^4 \rangle \simeq -\left\{ 128J^3[c_2 \left( 1 - \gamma_k^2 \right) \left( \zeta c_2 \left( \frac{3}{4}\gamma_k - \frac{3}{4} \right) - \frac{1}{4}c_0 \right) + c_0 c_1 \left( \frac{3}{4} - \frac{5}{4}\gamma_k + \frac{3}{4}\gamma_k^2 \right) + \zeta c_1 c_2 \left( \frac{13}{4} - \frac{15}{2}\gamma_k + \frac{17}{2}\gamma_k^2 \right) + \zeta c_2 \left( \frac{3}{2} - \frac{43}{3}\gamma_k + \frac{21}{4}\gamma_k^2 + \frac{5}{8} \cos k_x a \cos k_y a - 2\gamma_k^3 \right) \right]\nonumber
\]

\[
+ 16T_1 \nu_{eff} \left[ c_1 \left( 3 - 2\gamma_k^2 - \cos k_x a \cos k_y a \right) + \zeta T_2 \left( 7 - 12\gamma_k^2 + 5\gamma_k^3 \right) \right]
\]

\[
+ \frac{1 - \delta}{2} \left( 1 - 4\gamma_k^2 + 3\gamma_k^3 \right) + (\delta + \lambda) \left[ -\frac{9}{2} + 9\gamma_k - 3\gamma_k^2 - \frac{3}{2} \cos k_x a \cos k_y a \right]
\]

\[
+ 16t_{eff} J^2 T_1 c_0 \left( -\frac{39}{8} + \frac{31}{4}\gamma_k - \frac{23}{8}\gamma_k^2 \right) + c_2 \left( -\frac{85}{8} + \frac{93}{4}\gamma_k - \frac{101}{8}\gamma_k^2 \right)
\]

\[
+ c_1 \left( 16\gamma_k^2 - 35\gamma_k^3 + 25\gamma_k - \frac{9}{2} - \frac{3}{2} \cos k_x a \cos k_y a \right) + \frac{9}{16} \left( 1 - \frac{2}{7} \right) \left( \gamma_k - 1 \right)
\]

\[
+ 16t_{eff} J^2 c_1 \left( \frac{3}{4}\gamma_k - \frac{3}{4} \right) + T_2 \left( \frac{1}{4}T_0 + \frac{3}{4}T_0^2 - \lambda \right) \left( 2\gamma_k^2 - 3\gamma_k + 1 \right)
\]

\[
+ \frac{1 + \delta}{2} c_1 \left( 6\gamma_k^2 - \frac{45}{4}\gamma_k + \frac{21}{4} \right) + \frac{3}{4} \lambda \left[ (1 - T_0) - T_0^2 + \lambda T_0 \right] \left( \gamma_k - 1 \right)
\]

\[
+ \frac{1 + \delta}{2} c_2 \left( 2\gamma_k^2 - \frac{9}{2}\gamma_k + \frac{5}{2} \right) + c_1 T_0 \left( \frac{9}{4}\gamma_k^2 - \frac{9}{2}\gamma_k + \frac{1}{4} \right) + c_1 T_2 \left( \frac{11}{4}\gamma_k^2 - \frac{15}{2}\gamma_k + \frac{19}{4} \right)
\]

\[
+ c_2 T_0 \left( -2\gamma_k^2 + \frac{9}{2}\gamma_k - \frac{3}{2} \right) + \zeta T_2^2 \left( -4\gamma_k^2 + 6\gamma_k^2 + \frac{11}{4}\gamma_k - \cos k_x a \cos k_y a - \frac{15}{4} \right)
\]

\[
+ T_2 c_2 \left( 16\gamma_k^2 - 21\gamma_k^3 - \frac{5}{2}\gamma_k + \frac{15}{2} \right) + T_0 T_2 \left( 2\gamma_k^2 - \frac{9}{2}\gamma_k + \frac{9}{4} \right)
\]

\[
+ T_2 c_0 \left( -5\gamma_k^2 + \frac{9}{2}\gamma_k + \frac{1}{2} \right) + \zeta T_2^2 \left( -2\gamma_k^2 + 6\gamma_k^2 - \frac{19}{4}\gamma_k + \frac{3}{4} \right) \right)/\chi(k) \quad (15)
\]

(see Reference [40] for details). Note that in the expression for \( \langle \omega_k^4 \rangle \) the decoupling procedures were employed for the thermodynamic averages in spirit of papers by Hubbard and Jain [43], and by Kondo and Yamaji [44]. The averages with four operators are approximated, as usually, by products of two-operator correlation functions [39], however, multiplied now with the decoupling parameter \( \zeta \), e.g.: \( \langle S_i^\sigma S_j^\sigma S_{m}^\pi S_{n}^\pi \rangle \rightarrow \zeta \langle S_i^\sigma S_j^\sigma \rangle \langle S_{m}^\pi S_{n}^\pi \rangle \) and so on. This parameter may be fixed from the total moment sum rule, however, the uncertainty in the correlation length and the destruction of fraction of the Cu\(^{2+}\) moments by holes makes this restriction less rigorous and we fix \( \zeta \) from the comparison with experimental data.

### 2.3. Thermodynamic averages

To calculate the thermodynamic averages, we use the retarded Green’s functions formalism. The equation of motion for a retarded Green’s function \( \langle \langle A|B \rangle \rangle_\omega \) takes the form

\[
\omega \langle \langle A|B \rangle \rangle_\omega = \langle [A, B]_+ \rangle + \langle [A, H]|B \rangle \omega, \quad (16)
\]

where \( \langle ... \rangle \) denotes the thermal average. The standard relationship between correlation and Green’s function may be written as

\[
\langle BA \rangle = \frac{1}{2\pi i} \int d\omega f(\omega) \langle \langle A|B \rangle \rangle_\omega, \quad (17)
\]

where \( f(\omega) = [\exp (\omega/k_BT) + 1]^{-1} \) is the Fermi function; the contour encircles the real axis without enclosing any poles of \( f(\omega) \).
In general, Equation (16) cannot be solved exactly and one needs some sort of approximation. To evaluate the Green’s function $\langle[A, H]B\rangle_{\omega}$ in Equation (16), one uses a decoupling scheme originally proposed by Roth [46] for calculations on the Hubbard model. It can be shown that Roth’s method is essentially equivalent to the Mori-Zwanzig projection technique [47, 48] and is strongly related to the moments method as applied to the evaluation of the spectral density of the Green’s functions [49, 50]. Roth’s method has been studied by many authors [48, 51], and became a general method to treat approximately the quasiparticle spectrum of an interacting system. The reliability of the method has been demonstrated by comparison with the exact diagonalization results [51].

Roth’s method [46] implies that we seek a set of operators $A_n$, which are believed to be the most relevant to describe the one-particle excitations of the system of interest. Also, it is assumed that, in some approximation, these operators obey the relations [46]

$$[A_n, H] = \sum_m K_{nm} A_m,$$  \tag{18}

where the parameters $K_{nm}$ are derived through a set of linear equations

$$\langle[A_n, H], A_l \rangle_+ = \sum_m K_{nm} \langle[A_m, A_l^+] \rangle_+.$$  \tag{19}

Thus, it remains to define the operators $A_n$. Because, in the framework of the $t - J$ model, the quasiparticles are described by the Hubbard operators $X^0_k$, a set of operators $A_n$ contains only one operator $A = X^0_k$. Hence, the matrix $K_{nm}$ is diagonal and also contains one element $K = E_k^\sigma$, where $E_k^\sigma$ is the energy of of an electron with wave vector $k$ and spin projection $\sigma$. Consequently, Equations (18) and (19) become

$$[X^0_k, H] = E_k^\sigma X^0_k,$$  \tag{20}

$$\langle[X^0_k, H], X^{0\sigma}_k \rangle_+ = E_k^\sigma \langle[X^0_k, X^{0\sigma}_k] \rangle_+.$$  \tag{21}

In the 2D $t - J$ model, long-range order is absent at any finite temperature and hence, $E_k^\sigma$ does not depend on $\sigma$. Thus, we can replace $E_k^\sigma$ by $E_k$.

For our evaluations we need the thermal averages of the following types: $\langle X^{\sigma_0}_i X^{0\sigma}_j \rangle$ and $\langle X^{\sigma_0}_i X^{\sigma_0\sigma'}_j \rangle$. First, one should note, that in the absence of long-range order, $\langle X^{\sigma_0}_i \rangle$ does not depend on the site index and hence, $T_0 = \langle X^{\sigma_0}_i \rangle = \langle X^{\sigma_0}_i \rangle = (1 - \delta)/2$ and $c_0 = \langle S^z_i S^z_i \rangle = 1/4$.

The transfer amplitude between the first neighbours $T_1 = p I_1$ is given by

$$T_1 = p I_1 = -\frac{1}{2} \sum_{\rho} \langle X^{0\sigma}_i X^{0\sigma}_{i+\rho} \rangle$$  \tag{22}

and may be calculated using the spectral theorem

$$I_1 = -\sum_k \exp \left[ (E_k - \mu)/(k_B T) \right] + 1 \equiv \sum_k \gamma_k \rho_k.$$  \tag{23}

The latter equivalence has been obtained with the help of the identity $\sum_k \gamma_k = 0$. The sum (integral) over the wave vectors $k$ in the 2D Brilouin zone is normalized by its area, $(2\pi)^2$, which is omitted for brevity. The parameter $I_1$ in Equation (23) has been estimated in [52],

$$I_1 \approx \frac{4}{\pi} \left( 1 - e^{-\pi \delta} \right) - 2\delta, \quad \delta = \frac{\delta}{1 + \delta}.$$  \tag{24}
with an accuracy of a few percent over the whole region of \( \delta \) from 0 to 1. Here one should note that for very small \( \delta \) and low temperatures, \( I_1 \approx 2\delta \). Similarly, the transfer amplitude between the second neighbours, \( T_2 \), is given by

\[
T_2 = \frac{1}{z(z-1)} \sum_{\rho \neq \rho'} \langle X_i^{\sigma\sigma} X_i^{\sigma\sigma}_{i+\rho-\rho'} \rangle,
\]

\[
T_2 = \frac{p}{z(z-1)} \sum_k \left( 16\gamma_k^2 - 4 \cos k_x a \cos k_y a - 4 \right) \exp\left(\frac{(E_k - \mu)}{(k_B T)}\right) + 1
\]

\[
\equiv -\frac{p}{z(z-1)} \sum_k \left( 16\gamma_k^2 - 4 \cos k_x a \cos k_y a - 4 \right) f^h_k.
\]

(25)

\[
c_1 = \frac{1}{z} \sum_{\rho} \langle S_i^z S_{i+\rho}^z \rangle,
\]

\[
c_2 = \frac{1}{z(z-1)} \sum_{\rho \neq \rho'} \langle S_i^z S_{i+\rho-\rho'}^z \rangle,
\]

(26)

are the first and second neighbour spin-spin correlation functions respectively, the index \( \rho \) runs over nearest neighbours. The numerical values of \( c_1, c_2 \) have been calculated following the expressions as described in Reference [54].

For \( p \) we have

\[
p = (1 + \delta)/2,
\]

(27)

where \( \delta \) is the number of extra holes, due to doping, per one plane \( \text{Cu}^{2+} \), which can be identified with the Sr content \( x \) in \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \). The excitation spectrum of holes is given by

\[
E_k = 4t_{\text{eff}} f^h_k,
\]

(28)

where the hoppings, \( t \), are affected by electronic and AF spin-spin correlations \( c_1 \), resulting in effective values \([42, 52, 53]\), for which we set

\[
t_{\text{eff}} = \delta J/0.2
\]

(29)

to match the insulator-metal transition. The chemical potential \( \mu \) is related to \( \delta \) by

\[
\delta = p \sum_k f^h_k,
\]

(30)

where \( f^h_k = [\exp(-E_k + \mu)/k_B T + 1]^{-1} \) is the Fermi function of holes.

To obtain the thermodynamic averages of the type \( \langle X_i^{\sigma\sigma} X_i^{\sigma'\sigma'} \rangle \) it is convenient to make the following definitions

\[
\lambda = \lambda_{\tilde{\sigma}\tilde{\sigma}} = \frac{1}{z} \sum_{\rho} \langle X_i^{\tilde{\sigma}\tilde{\sigma}} X_{i+\rho}^{\tilde{\sigma}\tilde{\sigma}} \rangle,
\]

(31)

and

\[
\lambda_{\sigma\sigma'} = \frac{1}{z} \sum_{\rho} \langle X_i^{\sigma\sigma} X_{i+\rho}^{\sigma'\sigma'} \rangle.
\]

(32)

To obtain \( \lambda \) and \( \lambda_{\sigma\tilde{\sigma}} \) we use the two Green’s functions \([52]\)

\[
G^{(1)}_k(\omega) = \frac{1}{z} \sum_{\rho} \langle \langle X_k^{\tilde{\sigma}} | X_i^{\tilde{\sigma}0} X_{i+\rho}^{\tilde{\sigma}} \rangle \rangle \omega,
\]

(33)

\[
G^{(2)}_k(\omega) = \frac{1}{z} \sum_{\rho} \langle \langle X_k^{\tilde{\sigma}} | X_i^{\tilde{\sigma}0} X_{i+\rho}^{\tilde{\sigma}} \rangle \rangle \omega.
\]

(34)
Note that in the paramagnetic state, $\lambda_{\sigma\sigma} = \lambda_{\sigma\tilde{\sigma}}$ and $\lambda_{\sigma\tilde{\sigma}} = \lambda_{\tilde{\sigma}\sigma}$.

According to Equations (16) and (20), the equation of motion for $G^{(1)}_k(\omega)$ and $G^{(2)}_k(\omega)$ can be written as

\[
(\omega - E_k) G^{(1)}_k(\omega) = \frac{e^{ikr_i}}{\sqrt{N}} (1 - p - \lambda_{\sigma\sigma} + pI_1\gamma_k), \tag{35}
\]

\[
(\omega - E_k) G^{(2)}_k(\omega) = \frac{e^{ikr_i}}{\sqrt{N}} (1 - p - \lambda_{\tilde{\sigma}\tilde{\sigma}}), \tag{36}
\]

where $N$ is the number of sites. According to Equation (17):

\[
\lambda_{\tilde{\sigma}\tilde{\sigma}} = \frac{1}{2\pi i} \sum_k \frac{e^{-ikr_i}}{\sqrt{N}} \int d\omega f(\omega) G^{(1)}_k(\omega), \tag{37}
\]

\[
\lambda_{\sigma\sigma} = \frac{1}{2\pi i} \sum_k \frac{e^{-ikr_i}}{\sqrt{N}} \int d\omega f(\omega) G^{(2)}_k(\omega). \tag{38}
\]

Consequently, Equations (35) and (36) lead to a system of linear equations for $\lambda_{\tilde{\sigma}\tilde{\sigma}}$ and $\lambda_{\tilde{\sigma}\sigma}$ with the trivial solution

\[
\lambda = \lambda_{\tilde{\sigma}\tilde{\sigma}} = (1 - p)^2 - \frac{p^3}{2p - 1}I_1^2, \tag{39}
\]

\[
\lambda_{\sigma\tilde{\sigma}} = (1 - p - \lambda) \frac{1 - \delta}{1 + \delta} = (1 - p)^2 + \frac{(1 - p)p^2}{2p - 1}I_1^2. \tag{40}
\]

### 2.4. Decoupling procedures

We now describe the decoupling procedures for the thermodynamic averages performed following the papers by Hubbard and Jain [43] and by Kondo and Yamaji [44], and are performed in spirit of the self-consistent Born approximation (noncrossing approximation) [45].

The averages with the products of operators $X_i^{\sigma_0}X_j^{\sigma_0}$ are decoupled resulting in products of transfer amplitudes and the decoupling parameter $\zeta$,

\[
\langle X_i^{\sigma_0}X_j^{\sigma_0} X_m^{\tilde{\sigma}_0} X_j^{\tilde{\sigma}_0} \rangle \rightarrow \zeta \langle X_i^{\sigma_0}X_j^{\sigma_0} \rangle \langle X_m^{\tilde{\sigma}_0}X_j^{\tilde{\sigma}_0} \rangle. \tag{41}
\]

The four-spin correlation functions are approximated, as usually, by products of two-spin correlation functions [39], however, multiplied now with the decoupling parameter $\zeta$. Thus, we employ the decoupling procedures

\[
\langle S_i^{\sigma} S_r^{\tilde{\sigma}} S_m^{\tilde{\sigma}} S_j^{\sigma} \rangle \rightarrow \zeta \langle S_i^{\sigma} S_r^{\tilde{\sigma}} \rangle \langle S_m^{\tilde{\sigma}} S_j^{\sigma} \rangle, \tag{42}
\]

and

\[
\langle S_i^{\tilde{\sigma}} S_r^{\sigma} S_m^{\tilde{\sigma}} S_j^{\sigma} \rangle \rightarrow \zeta \langle S_i^{\tilde{\sigma}} S_r^{\sigma} \rangle \langle S_m^{\tilde{\sigma}} S_j^{\sigma} \rangle, \tag{43}
\]

for $i \neq r$ and $m \neq j$, whereas

\[
\langle S_r^{\sigma} S_r^{\tilde{\sigma}} S_m^{\tilde{\sigma}} S_j^{\sigma} \rangle \rightarrow 2\zeta \langle S_m^{\tilde{\sigma}} S_j^{\sigma} \rangle. \tag{44}
\]

The averages with the products of operators $X_i^{\sigma_0}X_r^{\tilde{\sigma}_0}$ between the nearest(next-nearest) neighbours and $(1 - X_m^{\tilde{\sigma}_0})(1 - X_j^{\sigma_0})$ are decoupled as follows:

\[
\langle X_i^{\sigma_0}X_r^{\tilde{\sigma}_0} (1 - X_m^{\tilde{\sigma}_0})(1 - X_j^{\sigma_0}) \rangle \rightarrow \zeta \langle X_i^{\sigma_0}X_r^{\tilde{\sigma}_0} \rangle (1 - X_m^{\tilde{\sigma}_0} - X_j^{\sigma_0} + X_m^{\tilde{\sigma}_0}X_j^{\sigma_0}) \tag{45}
\]

and so on.
The averages with spin and Hubbard operators are decoupled as follows:

\[
\langle X_i^{\sigma_0} X_j^{\sigma'} S_l^a S_r^\sigma \rangle \rightarrow \langle X_i^{\sigma_0} X_j^{\sigma'} \rangle \langle S_l^a S_r^\sigma \rangle,
\]

and with spin and density operators:

\[
\langle X_i^{\sigma} S_m^a S_r^\sigma \rangle \rightarrow \langle X_i^{\sigma} \rangle \langle S_m^a S_r^\sigma \rangle.
\]

The averages \(\langle X_i^{\sigma} X_j^{\sigma'} \rangle\) between the second neighbouring operators are decoupled simply by

\[
\langle X_i^{\sigma} X_j^{\sigma'} \rangle \rightarrow \langle X_i^{\sigma} \rangle \langle X_j^{\sigma'} \rangle,
\]

because an inspection of Equations (39) and (40) shows that the values of averages of these type between the first neighbours differ only slightly from \(\langle X_i^{\sigma} S_m^a S_r^\sigma \rangle\). Therefore, the averages between second neighbours in Equation (48) are thought as independent. In addition, because the averages \(\langle X_i^{\sigma} X_j^{\sigma'} \rangle\) between the first, in contrast with next-nearest, neighbours are calculated exactly, the averages like \(\langle X_i^{\sigma} X_j^{\sigma} X_j^{\sigma'} \rangle\) are decoupled in a way to avoid, where possible, the averages of the type as given in Equation (48).

2.5. Static susceptibility

In the present work we employ the static quantities that has been derived for both the carrier free and doped by charge carriers 2DHAF systems and work in the overall temperature range. The expression for static spin susceptibility is given by [54],

\[
\chi(k) = \frac{4|c_1|}{Jg-(g_+ + \gamma k)},
\]

and its structure is the same as in the isotropic spin-wave theory [16]. The meaning of \(g_+\) is clear: it is related to \(\xi\) via the expression \(\xi/a = 1/(2\sqrt{g_+ - T})\). Here we will use the doping and temperature dependence of \(\xi\) following the explicit formulation given in Reference [54].
To mimic the low temperature behavior of the correlation length we use the expression, as in Reference [52], resulting in effective correlation length $\xi_{eff}$, given by,

$$\xi_{eff}^{-1} = \xi_0^{-1} + \xi^{-1}.$$  

(50)

Here, in Equation (50), $\xi$ is affected by doped holes, in contrast with the Keimer et al. [13] empirical equation, where $\xi$ is given by the Hasenfratz-Niedermayer formula [15] and there was no influence of the hole subsystem on $\xi$. For strongly doped systems the expression for $\xi$ is much more complicated compared with simple relation $\xi/a \approx \sqrt{\frac{\delta}{k_BT}} \exp(2\pi\rho_S/k_BT)$, which is valid for carrier free or lightly doped systems [40, 54]. Thus from now on we replace $\xi$ by $\xi_{eff}$. In the best fit of $\xi_{eff}$ to NS data [13, 31] (see Figure 2) we use $\xi_0 = a/n_\xi \delta$, where $n_\xi$ is given in Table I. Whether its value follows from stripe ordering [55] or more exotic states [56] remains to be shown. The parameter $g_-$ in Equation (49) has been introduced in Reference [54] and its numerical values with doping are listed in Table I. The second neighbour spin-spin correlation function $c_2$ is related to $g_-$ as [54]: $g_- = \frac{2}{\delta}(1 + 30c_2)$.

3. Comparison with experiments and discussion

The results of the calculations are summarized in Table I. For brevity we consider here the cases: La$_{2-x}$Sr$_x$CuO$_4$ with $x = 0$, $x = 0.025$, $x = 0.035$, $x = 0.04$ and $x = 0.14$, and YBa$_2$Cu$_3$O$_{6.5}$ for which we accept [26] $\delta = 0.09$, and, particularly, $p = (1 + \delta/2)/2$, due to the bilayered structure that affects also $\langle \omega^2_k \rangle$, $\langle \omega^4_k \rangle$, and $\xi$.

Table 1. The calculated NN AF spin-spin correlation function $c_1 = \frac{1}{z} \sum_{\rho} \langle S^z_i S^z_{i+\rho} \rangle$, the parameter $g_-$, and the spin stiffness constant $\rho_S$ using the expressions and the procedure as described in References [54] and [52] in the $T \rightarrow 0$ limit together with the spin diffusion constant $D$ as calculated following References [40, 41] and the $n_\xi$ and $\zeta$ values as extracted from comparison with NS data.

| $\delta$ | $c_1$  | $g_-$  | $2\pi \rho_S / J$ | $D / J$ | $n_\xi$ | $\zeta$ |
|---------|--------|--------|-------------------|--------|--------|--------|
| 0.0     | -0.115215 | 4.1448 | 0.38              | 2.66   | -      | 1.8    |
| 0.04    | -0.1055 | 3.913  | 0.30              | 2.48   | 2      | 1.0    |
| 0.09    | -0.0851 | 3.46   | 0.24              | 3.5    | ~1.5   | 2.8    |
| 0.14    | -0.0657 | 3.034  | 0.15              | 6.5    | 1      | 4.0    |

Our results for $\chi''(k, \omega)$ agree with the basic relations known from general physical grounds for small wave vectors $q$ and frequency $\omega$ [8],

$$\chi''(q \sim 0, \omega \sim 0) \simeq \frac{2\chi_S \omega D q^2}{\omega^2 + (D q^2)^2},$$  

(51)

where

$$D = \lim_{q \rightarrow 0} \frac{1}{\pi q^2 F(q, 0)} = \lim_{q \rightarrow 0} \frac{1}{q^2} \sqrt{\pi \omega^2_q > 3 / (2 \omega^4_q >)}$$

is the spin diffusion constant. The Equation (51) for diffusive spin dynamics may be obtained from linear response theory, hydrodynamics and fluctuation-dissipation theorem. Using Equation (51) (or, equivalently, from Equation (9)) one may easy estimate the value of $q_0$ in the local maximum of the imaginary part of dynamic spin susceptibility $\chi''(q, \omega)$, which is given by

$$q_0^2 \simeq \omega / D.$$  

(52)
Figure 3. (Color online) Semilog-scale mesh of the calculated imaginary part of dynamic spin susceptibility $\chi''(k, \omega)$ in the Brillouin zone for (a) $T = 90$ K and $\delta = 0.09$ and (b) $T = 300$ K and $x = 0.04$. The cross on the vertical axis marks the value of $\chi''(q_0, \omega_{NQR} = 34 \text{ MHz})$ in its maximum at small wave vectors.

For typical value of the measuring NMR frequency, $\omega \approx 1 \text{ mK}$, $q_0 a \approx \pi \times 10^{-4}$. For extremely small $q \ll q_0$ with finite $\omega$ the imaginary part of dynamic spin susceptibility $\chi''(q, \omega)$ approaches zero: $\chi''(q_0 \gg q \to 0, \omega) \to 0$.

3.1. Inelastic neutron scattering

Figures 3-7 show the wave vector, frequency, doping and temperature dependence of $\chi''(k, \omega)$. We note that for all temperatures the form of $F(q, \omega)$ gives the elastic peak at $q = 0$ and $\omega = 0$. Figure 3 shows that for large $\omega$ the diffusive (small $k$) dynamics is negligible, the calculated $\chi''(k, \omega)$ for $\delta = 0.09$ is peaked at $Q = (\pi, \pi)$ for $\omega < 55$ meV and becomes incommensurate with a spin-wave like cone (symmetric ring of scattering) for $\omega > 55$ meV in agreement with high-energy NS studies [27].

Figure 4 shows $\chi''(Q, \omega)_{F}$ in the undamped approximation versus frequency and temperature.
Figure 5. Imaginary part of dynamic spin susceptibility $\chi''(k, \omega)$ versus $\omega$ (symbols: NS data for La$_{1.86}$Sr$_{0.14}$CuO$_4$ of the incommensurate peak from Reference [31]. The lines show the calculated $\chi''_F(Q = (\pi, \pi), \omega)$ in the undamped approximation.

Figure 6. The averaged over the Brillouin zone imaginary part of dynamic spin susceptibility $\chi''(\omega, T) = \int \chi''(q, \omega, T) dq$ versus $\omega/T$ (symbols: NS data for La$_{1.96}$Sr$_{0.04}$CuO$_4$ from Reference [13]. Solid lines show the calculated $\chi''_L(\omega, T)$) with Lorentzian, dashed lines show the calculated $\chi''_F(\omega, T)$ in the undamped spin-wave approximation.

The inset shows that $\chi''_F(Q, \omega)$ may not exhibit the sharp increase below $T_c$, in contrast with the predictions within the weak coupling theories [21, 22, 23]. Indeed, the more underdoped YBa$_2$Cu$_3$O$_{y\approx6.5}$ sample (controlled by $T_c$) with the smaller resonance frequency shows the smaller increase of $\chi''_F(Q, \omega)$ below $T_c$. Figure 5 shows the results of our calculations in spirit of undamped spin-wave picture of Kondo and Yamaji [44] and suggests that the damping of spin-wave like excitations affects $\chi''(k, \omega)$ noticeably in doped 2DHAF even at low temperatures. Noting that the relaxation shape function $F(k, \omega)$ can be understood within the spin-wave like [11] framework, $\omega_k^{SW} = 2 \int_0^\infty d\omega \omega F(k, \omega)$, the temperature and doping dependence of the damping of the spin-wave-like excitations may be studied further. Our results suggest that, in contrast with [18], the damping of spin-wave like excitations is, however, does not qualitatively affects $\chi''(k, \omega)$ even in the normal state of optimally doped high-$T_c$ cuprates. This may be caused by oversimplifications in [18] in the expression for susceptibility and simultaneous use of the temperature independent correlation length parameter, as indeed observed [13] only at $T < 400$ K in the lightly doped regime together with the numerical results that are valid solely in the $T > J/2 \approx 700$ K limit.

Figure 6 shows the averaged over the Brillouin zone and normalized imaginary part of dynamic spin susceptibility $\chi''(\omega, T)$ versus $\omega/T$. Both the undamped approximation and the Lorentzian form with damping for the imaginary spin susceptibility suggest the $\omega/T$ scaling for underdoped...
Figure 7. Temperature dependence of the dynamic structure factor $S(k, \omega)$, space and time Fourier transform of the spin-spin correlation function, as measured by neutron scattering in La$_{1.96}$Sr$_{0.04}$CuO$_4$ from top to bottom $\omega = 2$, $3$, $4.5$, $6$, $9$, $12$, $20$, $35$ and $45$ meV, respectively. The results of the calculations are given for $x = 0.04$ in both approaches: $\chi''(q, \omega, T)$ with damping (Lorentzian form) (solid lines) and in the approximation for undamped paramagnon-like excitations (dashed lines).

high-$T_c$ layered cuprates with a deviations in qualitative agreement with NS data [13].

Figure 7 shows the dynamic structure factor $S(k, \omega)$, space and time Fourier transform of the spin-spin correlation function, as measured by neutron scattering in La$_{1.96}$Sr$_{0.04}$CuO$_4$. At large and medium frequencies the agreement between theory and experiment is very good. At small $\omega$ both theoretical approaches have valuable deviations form experimental data. It was already mentioned in the original experimental reports that the behaviour at small $\omega$ in NS experiments deviates from universal curves and we therefore will compare the results of our calculations with NMR data in order to check also the absolute values of $\chi''(k, \omega)$ with considerably smaller $\omega$. 
Figure 8. The calculated plane oxygen nuclear spin-lattice relaxation rates \( \frac{17}{T_1} \) (lines) and the experimental data for \( La_{2-x}Sr_xCuO_4 \) as measured by NMR with \( x = 0.025 \) (triangles) and \( x = 0.035 \) (squares) from [60]. The experimental points have been rearranged with \( J = 1393 \) K. The results of the calculations in the undamped paramagnon-like excitations approximation with \( \omega = 2\pi \times 52 \) MHz are given for \( x = 0.035 \) by solid line and for \( x = 0.025 \) by dotted line. The contribution to \( \frac{17}{T_1} \) from spin diffusion for \( x = 0.035 \) with \( \omega = 2\pi \times 52 \) MHz is shown by upper dashed line and with \( \omega = 2\pi \times 81.4 \) MHz (14.1 T) by lower dashed line.

3.2. Plane copper and oxygen nuclear spin-lattice relaxation rates

The nuclear spin-lattice relaxation rate \( \frac{1}{T_1} \) is given by

\[
\frac{\alpha}{T_1} = \frac{2k_B T}{\omega_0} \sum_k \alpha F(k)^2 \chi''(k,\omega_0),
\]

where \( \omega_0 \ll T, J \) is the measuring NMR/NQR frequency. The quantization axis of the electric field gradient coincides with the crystal axis \( c \) which is perpendicular to CuO\(_2\) planes defined by \( a \) and \( b \). The wave vector dependent hyperfine formfactor for plane \( ^{63}\)Cu sites[58, 59] is given by \( ^{63}F(k)^2 = (A_{ab} + 4\gamma_k B)^2 \), where \( A_{ab} = 1.7 \cdot 10^{-7} \) eV and \( B = (1 + 4\delta) \cdot 3.8 \cdot 10^{-7} \) eV are the Cu on-site and transferred hyperfine couplings, respectively. The relation for \( B \) is used to match the weak changes with Sr doping [62]. For plane oxygen sites we use \( ^{17}F(k)^2 = 2C^2(1+\gamma_k) \), with \( C = 2.8 \cdot 10^{-7} \) eV.

We first estimate the value of contribution to \( 1/T_1 \) from small \( q \). A direct numerical integration over \( q \) is difficult, because \( \alpha F(q)^2 \chi''(q,\omega) \) has an extremely sharp peak at very small \( q_0 \). This requires an unattainably large number of points in numerical integration over the Brillouin zone. Expanding \( \chi''(q,\omega) \) around \( q_0 \) we obtain,

\[
\frac{\alpha}{T_1} = \frac{\alpha F(0)^2 k_B T \omega_0^2 \chi_S}{\pi \hbar D} \Lambda,
\]

where \( \Lambda \) depends on frequency through \( q_0 \). A simple and rough estimate gives

\[
\Lambda \sim \ln(1/q_0^2) \sim \ln(\text{const} \times J/\omega).
\]

This result explains the reason of the negligible shift of the oxygen \( \frac{17}{T_1} \) relaxation rate as measured by NMR at 9 Tesla (\( \omega_0 = 2\pi \times 52 \) MHz) and 14.1 Tesla (\( \omega_0 = 2\pi \times 81.4 \) MHz) that lies within the experimental accuracy [60]. One should note that \( \omega \) is much less than \( J = 1.8 \times 10^8 \) MHz, hence \( \ln(J/52 \text{ MHz})/\ln(J/81.4 \text{ MHz}) \approx 1.03 \). A sophisticated calculation
Figure 9. The calculated temperature dependencies of plane copper nuclear spin-lattice relaxation rate $^{63}(1/T_1) = 2W$ in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (data from [33]) for $x = 0.0$, $x = 0.04$ and for $x = 0.14$ by solid lines for $\chi''(q, \omega, T)$ with damping (Lorentzian form) and in the approximation for undamped paramagnon-like excitaions (dashed lines).

Figure 9. The calculated temperature dependencies of plane copper nuclear spin-lattice relaxation rate $^{63}(1/T_1) = 2W$ in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (data from [33]) for $x = 0.0$, $x = 0.04$ and for $x = 0.14$ by solid lines for $\chi''(q, \omega, T)$ with damping (Lorentzian form) and in the approximation for undamped paramagnon-like excitaions (dashed lines).

gives $\Lambda(35 \text{ MHz}) = 2.52$, $\Lambda(52 \text{ MHz}) = 2.44$, and $\Lambda(81.4 \text{ MHz}) = 2.37$. In view of the result that the spin diffusive contribution is $70 \%$, the relative shift of the measured $^{17}(1/T_1)$ will be $\approx 2 \%$, that lies within the experimental error (see Figure 8).

Figures 2 and 9 show the temperature dependencies of inverse correlation length and plane copper nuclear spin-lattice relaxation rate, $^{63}(1/T_1)$. Equations (11), (53), and Figures show that the temperature dependence of $^{63}(1/T_1)$ is governed by the temperature dependence of the correlation length and by the factor $k_BT$. At low $T$, where $\xi_{\text{eff}} \simeq \text{const}$, the plane copper $^{63}(1/T_1) \propto T$, as it should. At high $T$, the correlation length shows the weak doping dependence and behaves similarly to that of carrier free $\text{La}_2\text{CuO}_4$ and $^{63}(1/T_1)$ of doped samples behaves similarly to that of $\text{La}_2\text{CuO}_4$. Thus our results shown in Figures 2 and 8 suggest that the "pseudogap" effect seen with NMR in the high-$T_c$ cuprates is hidden in the correlation length that affects the observable quantities, and, generally, is in agreement with the conclusion based on the nearly AF Fermi liquid concept [61, 62] about the leading role of the correlation length in temperature and doping dependence of $1/T_1$.

4. Conclusion
In conclusion, we have presented a relaxation function theory for dynamic spin properties of layered copper high-$T_c$ in the normal state. The resonant feature, $\omega/T$ scaling, and spin-lattice relaxation at planar sites may be explained within the undamped spin-wave-like theory. We conclude that the resonant feature seen by NS may be caused not only by the feedback effect of the $d$-wave superconducting state. We discuss the influence of AF short range order (affected by stripes or, possibly, more exotic phases) on the dynamic quantities is discussed, possessing a reasonable agreement with the observations by means of neutron scattering and magnetic
resonance experiments in the underdoped high-$T_c$ layered copper oxides. At the same time, the future model for magnetic properties should match also a crossover from the localized spins picture (paramagnon-like excitation) to itinerant weak coupling theory with doping.

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