Magnetic properties of cuprate perovskites in the normal state

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Abstract

Normal-state magnetic properties of cuprate high-$T_c$ superconductors are interpreted based on the self-consistent solution of the $t$-$J$ model of Cu-O planes. The solution method retains the rotation symmetry of spin components in the paramagnetic state and has no preset magnetic ordering. The obtained solution is homogeneous. The calculated temperature and concentration dependencies of the magnetic susceptibility are close to those observed in experiment. These results offer explanations for the observed scaling of the static uniform susceptibility and for the changes in the spin correlation length, spin-lattice and spin-echo decay rates in terms of the temperature and doping variations in the spin excitation spectrum.

Key words: $t$-$J$ model, Cuprate perovskites, Magnetic properties

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Magnetic properties of cuprate perovskites have been extensively studied during the last years, both because of their unusual behavior and in the hope that they might provide insight into the physical origin of high-$T_c$ superconductivity [1,2,3,4,5,6,7]. Considerable progress has been made in this field with the use of phenomenological approaches, exact diagonalization of small clusters and – for heavily overdoped materials – in a RPA treatment. However, the basic issues of the magnetic behavior of underdoped cuprates, where strong electron correlations reveal themselves in full measure, have not yet been completely resolved. In this paper magnetic properties of underdoped cuprates in the normal state are investigated using the $t$-$J$ model of Cu-O planes and the method of Ref. [8] for calculating Green’s functions. This method has merits of retaining the rotation symmetry of spin components in the paramagnetic state and of the absence of any predefined magnetic ordering. Test calculations with this method for small clusters and for the undoped case demonstrated good agreement with the exact diagonalization and Monte Carlo results. For the

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considered 20×20 lattice and parameters of cuprates the calculated magnetic properties of the model appear to be close to those observed in these crystals. The results give an insight into mechanisms responsible for the unusual magnetic behavior of cuprate perovskites.

The Hamiltonian of the 2D $t$-$J$ model reads \[9\]

$$H = \sum_{nm\sigma} t_{nm} a_{n\sigma}^{\dagger} a_{m\sigma} + \frac{1}{2} \sum_{nm} J_{nm} \left( s_n^z s_m^z + s_n^{+1} s_m^{-1} \right),$$  \(1\)

where $a_{n\sigma} = |n\sigma\rangle\langle n0|$ is the hole annihilation operator, $n$ and $m$ label sites of the square lattice, $\sigma = \pm 1$ is the spin projection, $|n\sigma\rangle$ and $|n0\rangle$ are site states corresponding to the absence and presence of a hole on the site. For nearest neighbor interactions $t_{nm} = -t \sum_a \delta_{n,m+a}$ and $J_{nm} = J \sum_a \delta_{n,m+a}$ where $t$ and $J$ are the hopping and exchange constants and the four vectors $a$ connect nearest neighbor sites. The spin-$\frac{1}{2}$ operators can be written as $s_n^z = \frac{1}{2} \sum_\sigma \sigma |n\sigma\rangle\langle n\sigma|$ and $s_n^\sigma = |n\sigma\rangle\langle n,-\sigma|$.

The method suggested in Ref. \[8\] is based on Mori’s projection operator technique \[10\] which allows one to represent Green’s functions in the form of continued fractions and gives a way for calculating their elements. The residual term of the fraction can be approximated by the decoupling which reduces this many-particle Green’s function to a product of simpler functions. In this way we obtained the following self-energy equations for the hole $G(k\omega) = -i\theta(t)\langle\{a_{ko}(t), a_{ko}\}^\dagger\rangle$ and spin $D(k\omega) = -i\theta(t)\langle[s_k^z(t), s_{-k}^z]\rangle$ Green’s functions:

$$D(k\omega) = \frac{[4J\alpha(\Delta + 1 + \gamma_k)]^{-1} \Pi(k\omega) + 4JC_1(\gamma_k - 1)}{\omega^2 - \Pi(k\omega) - \omega_k^2},$$

$$G(k\omega) = \phi[\omega - \varepsilon_k + \mu - \Sigma(k\omega)]^{-1},$$  \(2\)

where $\gamma_k = \frac{1}{4} \sum_a \exp(ika)$, $\mu$ is the chemical potential, $\phi = \frac{1}{2}(1+x)$, $x$ is the hole concentration and

$$\omega_k^2 = 16J^2 \alpha |C_1|(1 - \gamma_k)(\Delta + 1 + \gamma_k),$$

$$\varepsilon_k = (4\phi t + 6C_1\phi^{-1} t - 3F_1\phi^{-1} J)\gamma_k.$$  \(3\)

The parameter of vertex correction $\alpha$, which improves the decoupling in the residual term, is set equal to its value in the undoped case, $\alpha = 1.802$. The parameter $\Delta$ which describes a gap in the spectrum of spin excitations at
\((\pi, \pi)\) [see Eq. (3)] is determined by the constraint of zero site magnetization \(\langle s^z_l \rangle = 0\) in the paramagnetic state. The constraint can be written in the form

\[
\frac{1}{2}(1 - x) = \frac{2}{N} \sum_k \int_0^\infty d\omega \coth\left(\frac{\omega}{2T}\right) B(k\omega),
\]

(4)

where \(B(k\omega) = -\pi^{-1} \text{Im} D(k\omega)\) is the spin spectral function, \(N\) is the number of sites and \(T\) is the temperature. Notice that in the considered 2D system the long-range antiferromagnetic ordering is destroyed at any nonzero \(T\) [11] and, as can be seen from the above formulas, at \(T = 0\) and \(x \gtrsim 0.02\). The value of \(x\) and the nearest neighbor correlations \(C_1 = \langle s^+_l s^-_{l+1} \rangle\) and \(F_1 = \langle a_l^\dagger a_{l+1} \rangle\) are defined from \(G(k\omega)\) and \(D(k\omega)\) in the usual way.

The self-energies in Eq. (2) read

\[
\text{Im} \Pi(k\omega) = \frac{16\pi t^2 J}{N} (\Delta + 1 + \gamma_k) \sum_{k'} (\gamma_k - \gamma_{k+k'})^2 
\times \int_{-\infty}^{\infty} d\omega' [n_F(\omega + \omega') - n_F(\omega')] A(k + k', \omega + \omega') A(k'\omega'),
\]

\[
\text{Im} \Sigma(k\omega) = \frac{16\pi t^2}{N \phi} \sum_{k'} \int_{-\infty}^{\infty} d\omega' [n_B(-\omega') + n_F(\omega - \omega')]
\times\left[\gamma_k-k' + \gamma_k + \text{sgn}(\omega')(\gamma_{k-k'} - \gamma_k)\sqrt{\frac{1 + \gamma_{k'}}{1 - \gamma_{k'}}}\right]^2
A(k - k', \omega - \omega') B(k'\omega'),
\]

(5)

where \(n_F(\omega) = [\exp(\omega/T) + 1]^{-1}\), \(n_B(\omega) = [\exp(\omega/T) - 1]^{-1}\) and \(A(k\omega) = -\pi^{-1} \text{Im} G(k\omega)\) is the hole spectral functions. The source of damping of spin excitations described by Eq. (5) is the decay into two fermions. Another source of damping, multiple spin excitation scattering, is considered phenomenologically by adding the small artificial broadening \(-2\eta \omega_k\), \(\eta = 0.02t\) to \(\text{Im} \Pi(k\omega)\).

The broadenings \(-\eta\) is also added to \(\text{Im} \Sigma(k\omega)\) to widen narrow lines and to stabilize the iteration procedure.

The same derivation for the transversal Green’s function gives \(\langle s^z_{k} | s^z_{k+1} \rangle = 2D(k\omega)\) indicating that the used approach retains properly the rotation symmetry of spin components in the paramagnetic state.

For low \(x\) and \(T\) the bandwidth of the dispersion \(\varepsilon_k\) is small in comparison with \(8t\), the bandwidth of uncorrelated electrons. This is a manifestation of the band narrowing in the antiferromagnetic surrounding.

Equations (2)–(5) form a closed set which was solved by iteration for the
parameters $t = 0.5$ eV, $J = 0.1$ eV corresponding to cuprates [12].

The frequencies of spin excitations satisfy the equation

$$\omega^2 - \text{Re}\Pi(k\omega) - \omega_k^2 = 0$$  \hspace{1cm} (6)$$

[see Eq. (2)]. For low $x$ and $T$ their dispersion is close to the dispersion of spin waves (see Fig. 1a). The main difference is the spin gap at $(\pi, \pi)$ the magnitude of which grows with $x$ and $T$ (Fig. 1b). In an infinite crystal this gap is directly connected with the spin correlation length $\xi$. Indeed, for large distances and low $T$ we find

$$\langle s^z_1 s^z_0 \rangle = N^{-1} \sum_k e^{ikl} \int_0^\infty d\omega \coth \left( \frac{\omega}{2T} \right) B(k\omega) \propto e^{iQl (\xi/|l|)^{1/2}} e^{-|l|/\xi},$$  \hspace{1cm} (7)$$

where $Q = (\pi, \pi)$ and $\xi = a/(2\sqrt{\Delta})$ with the intersite distance $a$. For low $x$ we found that $\Delta \approx 0.2x$ and consequently $\xi \approx a/\sqrt{T}$. This relation has been experimentally observed in La$_{2-x}$Sr$_x$CuO$_4$ [13].

As seen from Fig. 1b, with growing $x$ the spin excitation branch is destroyed in some region around the $\Gamma$ point – Eq. (6) has no solution for real $\omega$ due to large negative $\text{Re}\Pi(k\omega)$. For fixed $T$ the size of this region grows with $x$. In the considered model with rise of $T$ the branch is recovered in this region. This
is a consequence of the temperature broadening of the quasiparticle peak in
the hole spectrum. With this broadening $|\text{Re}\Pi|$ becomes smaller and Eq. (6)
has again a real solution.

An example of the variation of the spin correlations $C_{mn} = \langle s^z_l s^z_m \rangle$, $l = (m, n)$
with distance is given in Fig. 2. For large enough $x$ and $T$ the correlations decay exponentially with distance in the considered finite lattice. As mentioned,
the method used has no preset magnetic ordering. The character of the ordering is determined in the course of the calculations. As seen from Fig. 2,
only the short-range antiferromagnetic ordering was found in our calculations.
Stripes or other types of the phase separation were not revealed. Conceivably
such phase separations are not connected with the strong electron correlations described by the $t$-$J$ model.

The magnetic susceptibility is connected with the spin Green’s function (2) by
the relation $\chi^z(k\omega) = -4\mu_B^2 D(k\omega)$, where $\mu_B$ is the Bohr magneton. Experiments on inelastic neutron scattering give information on the susceptibility which can be directly compared with the calculated results. Such comparison is carried out in Fig. 3. $\text{YBa}_2\text{Cu}_3\text{O}_{7-y}$ is a bilayer crystal and the symmetry allows one to divide the susceptibility into odd and even parts. For the antiferromagnetic intrabilayer coupling the odd part can be compared with the calculated results. The oxygen deficiencies $y = 0.5$ and $0.17$ in the experimental data in Fig. 3 correspond to the hole concentrations $x = 0.05$ and $0.11$, respectively [14]. As seen from Fig. 3, the calculated data reproduce correctly the frequency dependence of the susceptibility, the values of the frequency for which $\text{Im}\chi(\pi, \pi)$ reaches maximum and their evolution with doping. The growth of the frequency of the maximum with $x$ reflects the respective increase of the spin gap. We notice also that the calculated temperature variation of the susceptibility is in good agreement with experiment. In absolute units the calculated maxima of $\text{Im}\chi(\pi, \pi)$ are $1.5 - 2$ times larger than the experimental values which is connected with some difference in decay widths of spin.

Fig. 2. Spin correlations along the diagonal [i.e., $l = (m, m)$] of the crystal for
$x = 0.12$. The respective temperatures are indicated near the curves.
Fig. 3. The imaginary part of the spin susceptibility for \( k = (\pi, \pi) \). Curves demonstrate calculated results for \( T = 0.02t \approx 116 \text{ K} \), \( x = 0.043 \) (a) and 0.08 (b). Squares are experimental results obtained in normal-state \( \text{YBa}_2\text{Cu}_3\text{O}_{7-y} \) at \( T = 100 \text{ K} \) for \( y = 0.5 \) (a) and 0.17 (b) [7].

Fig. 4. The uniform static spin susceptibility vs. \( T \).

Excitations.

The temperature dependence of the uniform static spin susceptibility \( \chi_0 = \chi(k \to 0, \omega = 0) \) is shown in Fig. 4. The calculated values lie in the range 2-2.6 eV\(^{-1}\) which is close to the values 1.9-2.6 eV\(^{-1}\) obtained for \( \text{YBa}_2\text{Cu}_3\text{O}_{7-y} \) [5]. The dependence \( \chi_0(T) \) has a maximum and its temperature \( T_m \) grows with decreasing \( x \). Analogous behavior is observed in cuprates for large enough \( x \) [1,3]. In Fig. 4 \( T_m \approx 600 \text{ K} \) which is close to \( T_m \) observed in \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) for comparable \( x \) [1]. As known, in the undoped antiferromagnet \( T_m \approx J \) [15]. On the high-temperature side \( \chi_0(T) \) tends to the Curie-Weiss dependence \( 1/T \). The decrease of \( \chi_0 \) below \( T_m \) is sometimes considered as the manifestation of the spin gap. In our opinion this statement is incorrect. For moderate \( x \) and \( T \) the long-wavelength part of the spin excitation spectrum does not feel the gap at \((\pi, \pi)\). For small but finite values of \( k \) \( \chi(k, 0) \propto \int_{-\infty}^{\infty} d\omega' B(k\omega')/\omega' \). The function \( B(k\omega') \) has a maximum which is shifted to lower frequencies and...
loses its intensity with increasing $T$ for such wave vectors. In the above integral the maximum is superimposed with the decreasing function $1/\omega'$ which finally leads to the nonmonotonic behavior of $\chi_0(T)$.

As seen from Fig. 4, the two curves for the different $x$ are very close in shape and can be superposed by scaling to the same values of maximum $\chi_0$ and $T_m$. Analogous scaling was observed in La$_{2-x}$Sr$_x$CuO$_4$ [1]. As follows from the above discussion, the source of this scaling is that holes and temperature fluctuations lead in a similar manner to the softening of the maximum in $B(k\omega')$ for long wavelengths.

The spin-lattice relaxation and spin-echo decay rates were calculated with the use of the equations [5]

\[
\frac{1}{\alpha T_1} = \frac{1}{2\mu_B^2 N} \sum_k \alpha F_{\beta}(k) \frac{\text{Im} \chi(k\omega)}{\omega}, \quad \omega \to 0,
\]

\[
\frac{1}{63T_{2G}} = \frac{0.69}{128\mu_B^4} \left\{ \frac{1}{N} \sum_k 63F_e^2(k) [\text{Re} \chi(k0)]^2 - \left[ \frac{1}{N} \sum_k 63F_e(k) \text{Re} \chi(k0) \right]^2 \right\},
\]

where the indices $\alpha$ and $\beta$ in the form factors $\alpha F_{\beta}(k)$ [5] indicate the nucleus type and the direction of the applied static magnetic field $H$, respectively. $\alpha = 63$ corresponds to Cu. The form factor $63F_e$ is the filter for the Cu spin-echo decay time $63T_{2G}$. Our calculated results and the respective experimental data are given in Fig. 5. The calculations reproduce satisfactorily main peculiarities of the temperature dependencies of the spin-lattice and spin-echo decay rates. The growth of $(63T_1)^{-1}$ with decreasing $x$ is connected with the increase of the spectral intensity of spin excitations near $(\pi, \pi)$ which make the main contribution to this rate. For the same $x$ $(63T_1)^{-1}$ is one order of magnitude larger than $(17T_1)^{-1}$, the spin-lattice relaxation rate at O sites (not shown here). This is a consequence of $\text{Im} \chi$ which is strongly peaked near $(\pi, \pi)$ and the form factors which test different $k$ regions [5]. The calculated spin-lattice relaxation rates are smaller than the experimental values due to the approximation made in the calculation of $D(k\omega)$ which somewhat underestimates $\text{Im} \chi$ at low frequencies.

For moderate $x$ with increasing $T$ the low-frequency region of $\text{Im} \chi(k \approx Q)$, $Q = (\pi, \pi)$ first grows due to the temperature broadening of the maximum in its frequency dependence and then decreases due to the temperature growth of the spin gap. It is the reason for the nonmonotonic behavior of $(63T_1)^{-1}$ in Fig. 5a. For moderate $x$ and low $T$ the magnitude of the spin gap is determined by the hole concentration and does not depend on $T$. This temperature range corresponds to the growth stage in Fig. 5a. The independence of the gap from
Fig. 5. The temperature dependencies of the spin-lattice relaxation and spin-echo decay rates at Cu sites. Open circles with right axes represent experimental results, filled circles with left axes are our calculations. (a,c) calculations for $H \parallel c$ and $x = 0.12$, measurements in YBa$_2$Cu$_3$O$_{6.63}$[3] ($x \approx 0.1$ [14]). (b) calculations for nonoriented configuration with $x = 0.043$, measurements in La$_{1.96}$Sr$_{0.04}$CuO$_4$ [2].

$T$ means that in this temperature range also $\xi$ does not depend on temperature which is a distinctive feature of the quantum disordered regime [4,5]. For temperatures above the maximum of $(^{63}T_1T)^{-1}$ we found that $^{63}T_1T/^{63}T_{2G} \approx \text{const}$ (see Fig. 5c) and $\xi^{-1} \propto \sqrt{\Delta} \propto T$ which points to the quantum critical $z = 1$ regime. These results are in agreement with the phenomenological treatment of experiment in YBa$_2$Cu$_3$O$_7-y$ carried out in Ref. [5] and the temperature of the maximum in Fig. 5a is close to $T_*$ of that work.

For small $x$ the spin gap grows with temperature starting from low $T$ and $(^{63}T_1T)^{-1}$ decreases monotonously, as seen in Fig. 5b. Due to the form-factor the region near $(\pi, \pi)$ does not contribute to $(^{17}T_1T)^{-1}$. There is a cardinal difference between the behavior of $\text{Im } \chi$ for $k \approx Q$ and away from $(\pi, \pi)$. Due to the spin gap in the former case the frequency of the maximum in $\text{Im } \chi(\omega)$ increases with $T$, while in the latter case, as mentioned above, it decreases. This frequency softening leads to the growth of the low-frequency $\text{Im } \chi$ and $(^{17}T_1T)^{-1}$ at low $T$ with their saturation for higher temperatures. Analogous behavior is observed in experiment [3].

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