Spin correlations and magnetic susceptibilities of lightly doped antiferromagnets

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Abstract

We calculate the spin correlation function and the magnetic longitudinal and transverse susceptibilities of a two-dimensional antiferromagnet doped with a small concentration of holes, in the t-J model. We find that the motion of holes generates spin fluctuations which add to the quantum fluctuations, the spin correlations decaying with the inverse of the spin distance, while increasing with doping as the critical hole concentration, where the long-range order disappears, is approached. Moreover, the longitudinal susceptibility becomes finite in the presence of doping, due to the strong damping effects induced by the hole motion, while the transverse susceptibility is renormalized by softening effects. Both the longitudinal and the transverse susceptibilities increase with doping, the former more significantly than the latter. Our results imply that doping
destroys the long-range order while local antiferromagnetic spin correlations persist. This is consistent with experiments on the doped copper oxide superconductors.

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Since their discovery,\textsuperscript{1} the copper oxide high-\(T_c\) superconductors have shown unusual magnetic characteristics, along with the unconventional transport properties.\textsuperscript{2} The undoped materials, e.g., La\(_2\)CuO\(_4\), are antiferromagnetic (AF) insulators, and doping, e.g., in La\(_{2-\delta}\)Sr\(_\delta\)CuO\(_4\), introduces holes, which are the charge carriers, in the spin lattice of the copper oxide planes. The CuO\(_2\) planes are described by a spin-1/2 Heisenberg antiferromagnet on a square lattice with moving holes that strongly interact with the spin array. A remarkable feature of the copper oxides is the strong dependence of their magnetic properties on the hole concentration \(\delta\). In previous work\textsuperscript{3–5} we studied the effects of doping on various magnetic properties, and showed that the motion of holes generates significant softening and damping of the spin excitations, leading, in particular, to the disappearance of the long-range AF order at a small hole concentration, due to the decay of spin-waves. We found that the staggered magnetization vanishes at a hole concentration well below the one for which the spin-wave velocity vanishes, or even the one for which all spin-waves become overdamped. This suggests that although the long-range order has disappeared, strong AF correlations persist, which allow the spin-wave excitations to exist. This is in agreement with experiments in the copper oxides, which show that, although the long-range order disappears, AF correlations persist up to fairly high doping, into the superconducting state.\textsuperscript{2,6–9} It is therefore of interest to study the spin correlations in these materials, because of their unusual behavior and their possible connection to high-\(T_c\) superconductivity.

In this work we use the \(t-J\) model to calculate the spin correlation function
of a two-dimensional antiferromagnet as a function of the hole concentration, which allows to investigate the local spin fluctuations, and also calculate the longitudinal and transverse magnetic susceptibilities, which reflect the global response of the system, accounting for the total spin fluctuations. We consider zero temperature and the low doping regime where the long-range AF order still exists. It is shown that the motion of holes generates spin fluctuations which add to the quantum fluctuations of the system, and increase with the hole concentration. Moreover, we find that the longitudinal spin susceptibility, which is zero in a pure Heisenberg antiferromagnet at zero temperature, becomes finite in the presence of doping, increasing significantly with the hole concentration, more pronouncedly than the corresponding transverse spin susceptibility.

We describe the copper oxide planes with the t-J model,

\[ H_{t-J} = -t \sum_{<i,j>,\sigma} (c_{i\sigma}^\dagger c_{j\sigma} + h.c.) + J \sum_{<i,j>} \left( \mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} n_i n_j \right), \tag{1} \]

where \( \mathbf{S}_i = \frac{1}{2} c_{i\sigma}^\dagger \sigma_{\alpha\beta} c_{i\beta} \) is the electronic spin operator, \( \sigma \) are the Pauli matrices, \( n_i = n_{i\uparrow} + n_{i\downarrow} \) and \( n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma} \). To enforce no double occupancy of sites, we use the slave-fermion Schwinger boson representation\(^{10}\) for the electronic operators \( c_{i\sigma} = f_i^\dagger b_{i\sigma} \), where the slave-fermion operator \( f_i^\dagger \) creates a hole and the boson operator \( b_{i\sigma} \) accounts for the spin, subject to the local constraint \( f_i^\dagger f_i + b_{i\uparrow}^\dagger b_{i\uparrow} + b_{i\downarrow}^\dagger b_{i\downarrow} = 2S \). For the undoped system, the model (1) describes a spin-1/2 Heisenberg antiferromagnet, exhibiting long-range Néel order at zero temperature. The Néel state is represented by a condensate of Bose fields \( b_{i\uparrow} = \sqrt{2S} \) and \( b_{i\downarrow} = \sqrt{2S} \), respectively, in the up and down sub-lattices, and the bosons \( b_i = b_{i\downarrow} \) and \( b_j = b_{j\uparrow} \) are then spin-excitation operators on the Néel
background. After a Bogoliubov-Valatin transformation on the boson Fourier transform, \( b_k = u_k \beta_k + v_k \beta_k^\dagger \), where 
\[
u_k = -\text{sgn}(\gamma_k) \left[(1 - \gamma_k^2)^{-1/2} - 1\right]^{1/2},\]
with 
\[
\gamma_k = (\cos k_x + \cos k_y)/2,
\]
we arrive at the effective Hamiltonian

\[
H = -\frac{1}{\sqrt{N}} \sum_{q,\mathbf{k}} f_q f_{\mathbf{q}-\mathbf{k}}^\dagger \left[V(\mathbf{q}, -\mathbf{k}) \beta_{-\mathbf{k}} + V(\mathbf{q} - \mathbf{k}, \mathbf{k}) \beta_{\mathbf{k}}^\dagger\right] + \sum_{\mathbf{k}} \omega_{\mathbf{k}} \beta_{\mathbf{k}}^\dagger \beta_{\mathbf{k}},
\]
where, 
\[
V(\mathbf{q}, \mathbf{k}) = zt(\gamma_{\mathbf{q}} u_{\mathbf{k}} + \gamma_{\mathbf{q}+\mathbf{k}} v_{\mathbf{k}})
\]
represents the interaction between holes and spin-waves resulting from the motion of holes with emission and absorption of spin-waves, 
\[
\omega_{\mathbf{k}} = (zJ/2)(1 - \gamma_k^2)^{1/2}
\]
is the dispersion for spin-waves in the undoped antiferromagnet, and \( z \) is the lattice coordination number (\( z = 4 \)), \( N \) the number of sites in each sub-lattice. The sums are performed in the first Brillouin zone of an antiferromagnet on a square lattice.

The magnetic properties are calculated in terms of the spin-wave Green’s functions

\[
D^{\mu\nu}(\mathbf{k}, t - t') = -i < \mathcal{T} \beta_{\mathbf{k}}(t) \beta_{\mathbf{k}}^\dagger(t') >
\]

\[
D^{\mu - \nu}(\mathbf{k}, t - t') = -i < \mathcal{T} \beta_{\mathbf{k}}^\dagger(t) \beta_{-\mathbf{k}}(t') >
\]

\[
D^{\mu - \nu}(\mathbf{k}, t - t') = -i < \mathcal{T} \beta_{\mathbf{k}}(t) \beta_{-\mathbf{k}}(t') >
\]

\[
D^{\mu + \nu}(\mathbf{k}, t - t') = -i < \mathcal{T} \beta_{\mathbf{k}}^\dagger(t) \beta_{\mathbf{k}}^\dagger(t') >,
\]
where \(< \ldots \rangle \) represents the average over the ground state. The spin-wave Green’s functions verify the Dyson equations

\[
D^{\mu\nu}(\mathbf{k}, \omega) = D^{\mu\nu}_0(\mathbf{k}, \omega) + \sum_{\alpha\gamma} D^{\mu\alpha}_0(\mathbf{k}, \omega) \Pi^{\alpha\gamma}(\mathbf{k}, \omega) D^{\gamma\nu}(\mathbf{k}, \omega),
\]
with \( \mu, \nu, \alpha, \gamma = \pm \). The free Green’s functions are 
\[
D^{\mu\nu}_0(\mathbf{k}, \omega) = 1/(\omega - \omega_0^{\mu} + i\eta),
\]
\[
D^{\mu - \nu}_0(\mathbf{k}, \omega) = 1/(-\omega - \omega_0^{\mu} + i\eta),
\]
(\( \eta \to 0^+ \)), 
\[
D^{\mu - \nu}_0(\mathbf{k}, \omega) = D^{\mu + \nu}_0(\mathbf{k}, \omega) = \]
0. $\Pi^{\alpha\gamma}(k, \omega)$ are the self-energies generated by the interaction between holes and spin-waves, which we calculate in the self-consistent Born approximation (SCBA). This corresponds to considering only "bubble" diagrams with dressed hole propagators, describing the decay of spin-waves into "particle-hole" pairs. The spin-wave self-energies take the form

$$\Pi^{\alpha\gamma}(k, \omega) = \frac{1}{N} \sum_{q} U^{\alpha\gamma}(k, q) \left[ Y(q, -k; \omega) + Y(q - k, k; -\omega) \right],$$

(3)

with $U^{\alpha\gamma}(k, q) = V(q, -k)V(q-k, k)$, $U^{\alpha\gamma}(k, q) = V(q-k, k)^2$, $U^{\alpha\gamma}(k, q) = V(q, -k)^2$ and

$$Y(q, -k; \omega) = \int_{0}^{+\infty} d\omega' \int_{-\infty}^{0} d\omega'' \rho(q, \omega') \rho(q-k, \omega'') \frac{\omega + \omega'' - \omega' + i\eta}{\omega + \omega'' - \omega' + i\eta}.$$  

The SCBA provides a spectral function for the holes, $\rho(q, \omega)$, which is composed of a coherent quasiparticle peak with weight $a_0 \simeq (J/t)^{2/3}$ and dispersion $\epsilon_q \simeq \epsilon_{\min} + (q - q_i)^2/2m$, with effective mass $m \simeq 1/J$, the Fermi surface for the holes consisting of pockets, of radius $q_F = \sqrt{\pi\delta}$, located at $q_i = (\pm \frac{\pi}{2}, \pm \frac{\pi}{2})$ in the Brillouin zone, and an incoherent continuum taking the approximate form $h\theta(|\omega| - zJ/2)\theta(2zt + zJ/2 - |\omega|)$, with $h \simeq (1 - a_o)/2zt$. We calculated the self-energies to lowest order in the hole concentration $\delta$.

The spin correlation function is defined as

$$C(r) = \frac{1}{2N} \sum_{j} \left( \langle \mathbf{S}_j \cdot \mathbf{S}_{j+r} \rangle - \langle \mathbf{S}_j \rangle \cdot \langle \mathbf{S}_{j+r} \rangle \right),$$

(4)

where the sum runs over all lattice sites. Writing the spin operators, $S_j^z$, $S_j^x = (S_j^+ + S_j^-)/2$, $S_j^y = (S_j^+ - S_j^-)/2i$, in terms of the electron operators, one has, $S_j^z = (c^\dagger_{j\uparrow} c_{j\uparrow} - c^\dagger_{j\downarrow} c_{j\downarrow})/2$, $S_j^+ = c_{j\uparrow}^\dagger c_{j\downarrow}$, $S_j^- = c_{j\downarrow}^\dagger c_{j\uparrow}$, which, using the Schwinger...
boson representation and the boson condensation associated with the Néel state, leads to 
\[ S_z^j = (1 - \delta)(1/2 - b_j^\dagger b_j), \quad S_j^+ = (1 - \delta)b_j, \quad S_j^- = (1 - \delta)b_j^\dagger, \]
for the up sub-lattice, and 
\[ S_z^j = -(1 - \delta)(1/2 - b_j^\dagger b_j), \quad S_j^+ = (1 - \delta)b_j^\dagger, \quad S_j^- = (1 - \delta)b_j, \]
for the down sub-lattice, having done the approximation \( f_j f_j^\dagger = 1 - \delta \). In terms of the spin-excitation boson operators one has

\[ C(r) = (1 - \delta)^2 \frac{1}{4N} \left[ \sum_{j \in \mathbb{Z}^2} \left( \langle b_j b_{j+r}^\dagger \rangle + \langle b_{j+r} b_j^\dagger \rangle + 2 \langle b_j b_{j+r} b_{j+r}^\dagger \rangle \right) - 2 \langle b_j b_{j+r} \rangle \langle b_{j+r} b_j^\dagger \rangle \right]. \]

After Fourier transform and the Bogoliubov-Valatin transformation, we make the mean-field decoupling \( \langle ABCD \rangle \approx \langle AB \rangle \langle CD \rangle + \langle AC \rangle \langle BD \rangle + \langle AD \rangle \langle BC \rangle \). This allows to express the correlation function (5) in terms of the spin-wave Green’s functions in the form

\[ C(r) = (-1)^{x+y}(1 - \delta)^2 \left( \frac{1}{N} \sum_k (u_k^2 + v_k^2) \cos(k \cdot r) \right) \times \left[ 1 - \int_0^{\infty} \frac{d\omega_1}{2\pi} \left( 2\text{Im}D^{++}(\mathbf{k}, \omega) + 4u_k v_k \text{Im}D^{--}(\mathbf{k}, \omega) \right) \right] \]
\[ + \frac{4}{N^2} \sum_{\mathbf{k}_1, \mathbf{k}_2} \int_0^{\infty} \frac{d\omega_1}{2\pi} \int_0^{\infty} \frac{d\omega_2}{2\pi} \cos((\mathbf{k}_1 - \mathbf{k}_2) \cdot r) \]
\[ \times \left[ (u_{\mathbf{k}_1}^2 - (u_{\mathbf{k}_1}^2 + v_{\mathbf{k}_1}^2)\text{Im}D^{++}(\mathbf{k}_1, \omega_1) - 2u_{\mathbf{k}_1} v_{\mathbf{k}_1} \text{Im}D^{--}(\mathbf{k}_1, \omega_1) \right] \]
\[ \times \left[ (u_{\mathbf{k}_2}^2 - (u_{\mathbf{k}_2}^2 + v_{\mathbf{k}_2}^2)\text{Im}D^{++}(\mathbf{k}_2, \omega_2) - 2u_{\mathbf{k}_2} v_{\mathbf{k}_2} \text{Im}D^{--}(\mathbf{k}_2, \omega_2) \right] \}, \]

where \( r = (x, y) \). The pre-factor of \((-1)\) arises when the correlation is between sites on different sub-lattices.
To lowest order in the hole concentration, \( \delta \), we obtain for the correlation function (6), the expression

\[
C_0(r) = (1 - \delta)^2 \left[ C_o(r) + C_\delta(r) \right]
\]

where

\[
C_0(r) = \frac{1}{2N} \sum_k \left( u_k^2 + v_k^2 \right) \cos(k \cdot r)
\]

is the correlation function for a pure Heisenberg antiferromagnet, accounting for the quantum fluctuations, and

\[
C_\delta(r) = - \left[ 1 + \frac{1}{N} \sum_k \left( u_k^2 + v_k^2 \right) \cos(k \cdot r) \right] \frac{1}{N} \sum_k \cos(k \cdot r)
\]


times

\[
 \times \left[ u_k v_k \frac{\text{Re}\Pi(k, \omega^2_k)}{\omega^2_k} - 2u_k v_k \int_0^{+\infty} \frac{d\omega}{\pi} \frac{\text{Im}\Pi(k, \omega)}{(\omega^2 - (\omega^2_k)^2)} \right]
\]

contains the effect of doping on the spin correlations associated to the hole motion; the pre-factor \( (1 - \delta)^2 \) corresponds to spin dilution, being negligible in the low doping regime considered. In Fig.1 we present the correlation function \( C(r) \), Eq.(7), calculated for two different directions, \( x = y \) and \( y = 0 \), in the case of \( \delta = 0.02 \), and the pure case, \( \delta = 0.0 \). One sees that the spin correlations are independent of the spatial direction, a result that is verified at any doping.

In Fig.2 we plot the correlation function as a function of the spin distance \( r \), for various hole concentrations. We observe that \( C(r) \) increases with doping, and decays, at large distances, as \( 1/r \) (Fig.2 inset), both in the pure and in
the doped cases. One can describe the behavior of $C(r)$ at large distances as

$$C(r) = A(\delta)/r,$$

where $A(\delta) = A_o + B\delta^\alpha$, with $A_o = 1/\sqrt{2\pi}$ and $\alpha = 0.42$. $A(\delta)$ contains the doping dependence, which is represented in Fig.3. The dominant contribution to $C_\delta(r)$, at large $r$, comes from the imaginary part of the spin-wave self-energies, which depend on the hole concentration essentially as $\sqrt{\delta}$. In Fig.4 we compare the increase of $C(r)$ with the hole concentration $\delta$ at fixed small $r$ (Fig.4a) and large $r$ (Fig.4b). The decay of $C(r)$ with $1/r$ was expected for the undoped case since in a two dimensional antiferromagnet at zero temperature the correlation length is infinite. One also expected $C(r)$ to increase with doping since the motion of holes generates spin fluctuations which eventually lead to the destruction of the long-range AF order at a finite critical concentration $\delta_c$. In previous work, we found that the staggered magnetization vanishes at a small critical concentration (e.g. $\delta_c \simeq 0.07$ for $t/J = 3$), while the long-wavelength spin excitations remain well defined up to a higher hole concentration ($\delta^* \simeq 0.17$ also for $t/J = 3$). Here, we find that the doping does not qualitatively change the behavior of $C(r)$ with $r$, as compared to the pure case, which reflects the robustness of the local AF order in the doped material. Spin correlations in the copper oxides were studied before, both experimentally and theoretically, but in a higher doping regime where the long-range AF order has already disappeared. In this regime the spin correlations decrease with increasing doping, as the system moves away from the critical hole concentration.

In the presence of long-range AF order one distinguishes a longitudinal and
a transverse susceptibility. The longitudinal spin susceptibility is defined as

$$\chi_{\parallel} = \chi_{\parallel}(\mathbf{k} = 0, \omega = 0),$$  

where the dynamical susceptibility is given by

$$\chi_{\parallel}(\mathbf{k}, \omega) = i \int_{0}^{+\infty} dt e^{i\omega t} \langle [S^z(\mathbf{k}, t), S^z(-\mathbf{k}, 0)] \rangle.$$ 

In terms of the spin-wave Green’s functions one has

$$\chi_{\parallel} = \lim_{\mathbf{k} \to 0} \frac{1}{N} \sum_{\mathbf{k}_1} \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \left[ 2u_{\mathbf{k}_1} v_{\mathbf{k}_1} u_{\mathbf{k}_1} v_{\mathbf{k}_1} - u_{\mathbf{k}_1}^2 v_{\mathbf{k}_1}^2 - v_{\mathbf{k}_1}^2 u_{\mathbf{k}_1}^2 \right]$$

$$\times D^{+}\left(\mathbf{k}_1, \omega\right) D^{+}\left(\mathbf{k}_1 - \mathbf{k}, -\omega\right)$$

which to lowest order in the hole concentration gives

$$\chi_{\parallel} = \frac{1}{N} \sum_{\mathbf{k}} \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \frac{\text{Im}\Pi^{+\pm}(\mathbf{k}, \omega)}{(\omega - \omega_{\mathbf{k}}^\mp)^3}. \quad (11)$$

The transverse spin susceptibility is defined by

$$\chi_{\perp} = \chi_{\perp}(\mathbf{k} = 0, \omega = 0),$$  

where

$$\chi_{\perp}(\mathbf{k}, \omega) = i \int_{0}^{+\infty} dt e^{i\omega t} \langle [S^x(\mathbf{k}, t), S^x(-\mathbf{k}, 0)] \rangle.$$ 

In terms of the spin-wave Green’s functions the transverse spin susceptibility is expressed as

$$\chi_{\perp} = -\lim_{\mathbf{k} \to 0} \left( \frac{1 - \gamma_{\mathbf{k}}}{1 + \gamma_{\mathbf{k}}} \right)^{1/2} \left[ \text{Re}D^{+\mp}(\mathbf{k}, 0) + \text{Re}D^{+\mp}(\mathbf{k}, 0) \right],$$

which, to lowest order in the hole concentration $\delta$, is given by

$$\chi_{\perp} = \lim_{\mathbf{k} \to 0} \frac{1}{zJ(1 + \gamma_{\mathbf{k}})} \left[ 1 - \frac{2}{zJ(1 - \gamma_{\mathbf{k}}^2)^{1/2}} \left[ \text{Re}\Pi^{+\mp}(\mathbf{k}, 0) + \text{Re}\Pi^{+\mp}(\mathbf{k}, 0) \right] \right]. \quad (13)$$
We found that $\chi_\perp$ takes the simple form

$$\chi_\perp = Z \chi^0_\perp$$

where $\chi^0_\perp = 1/(2zJ)$ is the transverse spin susceptibility for a pure Heisenberg antiferromagnet, and $Z \chi = 1 + 4\delta a^2 \left(t/J\right)^2$ is a renormalization factor.

Comparing Eqs. (11) and (13) one sees that the motion of holes influences the longitudinal and transverse susceptibilities in different ways, the former is produced by the imaginary part of the self-energy, while the latter is renormalized by the real part of the self-energies. In a pure Heisenberg antiferromagnet the longitudinal susceptibility is zero. However, with doping $\chi_\parallel$ acquires a finite value due to the decay of spin-waves into "particle-hole" pairs, generated by hole motion. The renormalization of $\chi_\perp$ reflects a softening of the spin coupling induced by the hole motion. In Figs. 5 and 6 we plot the longitudinal, Eq.(11), and the transverse, Eq.(13), susceptibilities as a function of the hole concentration for $t/J = 3, 4$, in the doping range where the long-range AF order exists, in the approach considered. We find that both susceptibilities increase with doping, though the longitudinal one is far more sensitive to the hole concentration than the transverse one. The transverse susceptibility reflects the stiffness of the antiferromagnetic lattice. In contrast, the longitudinal susceptibility is set by the strong damping effects, which are also responsible for the disappearance of the long-range AF order at low doping. When the long-range order is broken, the susceptibility of the system should be essentially given by

$$\chi = \frac{1}{3} \chi_\parallel + \frac{2}{3} \chi_\perp,$$

with the longitudinal susceptibility providing an important contribution. Also, in the ceramic samples whose crystal axis are randomized
the susceptibility $\chi$ is given by an average of the susceptibilities for the three directions. An increase of the spin susceptibility with doping has in fact been observed experimentally.$^{21-24}$

In summary, we studied the effects of hole motion on the spin correlation function and the magnetic longitudinal and transverse susceptibilities of a two-dimensional antiferromagnet doped with a small concentration of holes. We found that the spin fluctuations increase with doping, the spin correlations decaying with the inverse of the spin distance, which indicates that the local AF correlations remain quite robust. Furthermore, we show that the longitudinal magnetic susceptibility acquires a finite value in the presence of doping, due to the strong damping effects generated by the hole motion, while the transverse magnetic susceptibility is renormalized. Both susceptibilities show a significant increase with doping, which is however more pronounced in the longitudinal one. Our results imply that doping destroys the long-range AF order while local spin correlations persist. This is consistent with experimental observations in the copper oxide high-$T_c$ superconductors.

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Figure Captions:

FIG. 1. Correlation function $C(r)$ vs spin distance $r$, at the hole concentration $\delta = 0.02$, for the directions $x = y$ (open circles) and $y = 0$ (diamonds) on the square lattice, with $t/J = 3$. Inset: $C(r)$ vs $r$, in the pure antiferromagnet ($\delta = 0.0$).

FIG. 2. Correlation function $C(r)$ vs spin distance $r$, $x = y$, for various hole concentrations $\delta$, with $t/J = 3$. Inset: $C(r)$ vs $1/r$ for, large $r$.

FIG. 3. Spin correlation amplitude $A(\delta)$, at large spin distances, vs hole concentration $\delta$, with $t/J = 3$.

FIG. 4. Correlation function $C(r)$ vs hole concentration $\delta$, with $t/J = 3$, for fixed $r$, $x = y$. a: in the range of small $r$. b: in the range of large $r$.

FIG. 5. Longitudinal susceptibility $\chi_{\parallel}$ as a function of doping $\delta$, for $t/J = 3$ (open circles) and $t/J = 4$ (diamonds).

FIG. 6. Transverse susceptibility $\chi_{\perp}$ as a function of doping $\delta$, for $t/J = 3$ (open circles) and $t/J = 4$ (diamonds).
Fig. 1
Fig. 2
Fig. 3
Fig. 4
Fig. 5
Fig. 6