Proc. Estonian Acad. Sci. Phys. Math., 1995, 44, 2/3, 286–296
https://doi.org/10.3176/phys.math.1995.2/3.18

MAGNETIC EXCITATIONS IN LOW-DOPED CuO_2 LAYERS

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Received 22 September 1994, accepted 17 April 1995

Abstract. The copper spins of undoped CuO_2 layers in the high-T_c cuprates are ordered antiferromagnetically. For small dopings this antiferromagnetic (AF) order is preserved while spin-polarized clusters are formed. With an equation of motion technique for Green's functions the influence of a local-spin-polarized perturbation on the AF magnon spectrum is calculated. By using this method temperature-dependent magnetization and susceptibility are obtained for different doping concentrations. It turns out that localized dispersionless states split from AF magnon bands.

Key words: high-T_c cuprates, low doping, magnetic susceptibility.

INTRODUCTION

In the last years ever increasing evidence has been found for the existence of low-energetic magnetic excitations within high-T_c materials [1, 2]. At the same time, theoretical models suggesting an electronically driven phase separation have got increasing attention [3, 4]. In an undoped case (e.g. La_2CuO_4, $YBa_2Cu_3O_6$) high-T_c cuprates are antiferromagnetic (AF) insulators with remarkably high Neel temperatures ($LaCu_2O_4$: $T_N \approx$ 250 K). This means that copper spins in CuO_2 layers are ordered antiferromagnetically. If one dopes the systems (by introducing O, Sr or other divalent atoms), holes are introduced into the layers. Already at very low doping concentrations, e.g. $La_{2-\delta}Sr_\delta CuO_4$, $\delta \approx 0.03$, the long-range AF order is destroyed, while some short-range AF correlations are still observable. For higher hole concentrations ($\delta > 0.08$) the systems become conducting and below T_c, superconducting. In this work we lay emphasis on low doping concentrations of $\delta < 0.05$.

The basic idea of [4] is as follows:
A single hole introduced into a CuO_2 layer will form a magnetic polaron (see Fig. 1) with a high effective mass. The binding energy of such a polaron is $E \approx 0.4$ eV, its size will be 5–8 Cu sites, while the polarization at Cu sites is approximately 80% and it decreases with further doping. The interaction of such polarons is very small, as follows from detailed mean field calculations by using the three-band Hubbard model [5, 6]. On further doping this leads to a phase separation into a hole-rich phase formed by magnetic polarons and a hole-poor phase that is responsible for the remaining antiferromagnetism.

From this picture we conclude that the magnetic susceptibility of the system for the low doping (still insulating) range is given by three parts:

1. A spin wave part that is responsible for the increase towards the Neel peak;

2. A Curie part resulting from the spins of the magnetic polarons, leading to a divergence at zero temperature;

3. The influence of the perturbation of the AF magnon spectrum which gives a weakly temperature-dependent part calculated here.

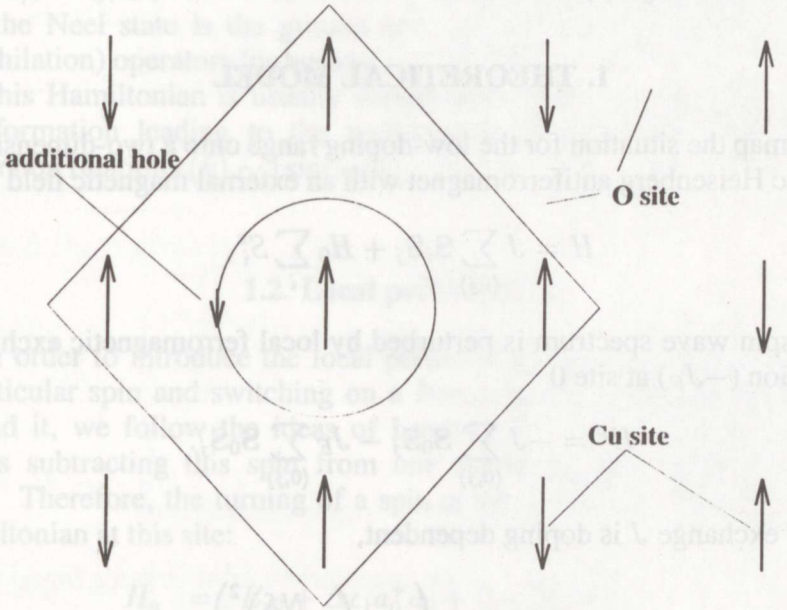

Fig. 1. A schematic sketch of the smallest magnetic polaron formed in the AF-ordered CuO_2 plane via doping with an additional hole. The arrows indicate the main direction of spins only.

Figure 2 shows the measurements by Johnston et al. [7]. It can be seen that this picture fits at least qualitatively the experimental data. A decrease of the Neel peak with the increasing doping is observed belonging to part 1. The increase of the Curie signal for low temperatures that gets more important with higher doping is due to part 2.

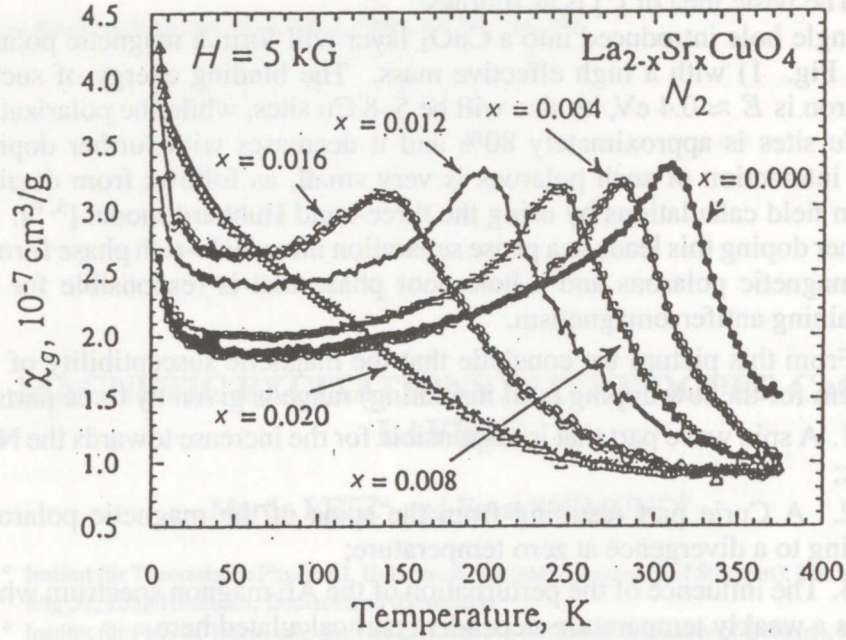

Fig. 2. The measured magnetic susceptibility χ_g versus temperature for various doping concentrations according to [7].

1. THEORETICAL MODEL

We map the situation for the low-doping range onto a two-dimensional isotropic Heisenberg antiferromagnet with an external magnetic field H_0

$$H = J \sum_{\langle i,j \rangle} \mathbf{S}_i \mathbf{S}_j + H_0 \sum_i S_i^z, \tag{1}$$

whose spin wave spectrum is perturbed by local ferromagnetic exchange interaction $(-J_F)$ at site 0

$$H^p = -J \sum_{\langle 0,j \rangle} \mathbf{S}_0 \mathbf{S}_j - J_F \sum_{\langle 0,j \rangle} \mathbf{S}_0 \mathbf{S}_j. \tag{2}$$

The AF exchange J is doping dependent,

$$J = J(\delta) = J_0 \left(1 - \left(\alpha \frac{N\delta}{2}\right)^2\right), \tag{3}$$

since the region of $N\delta$ has no AF exchange any more. N is the size of the cluster, and $\alpha > 1$ reflects the circumstance that due to fluctuations the magnetic polaron destroys the antiferromagnetism of a larger region. Factor $1/2$ results from two spin directions and $()^2$ indicates that the oxygen site enters the AF superexchange twice. According to [8], $J_0 \approx \frac{4t^4}{(U-\epsilon)^3} \approx$ ≈ 32 meV and $J_F \approx \frac{t^2}{U-\epsilon}\frac{1}{4} \approx 50$ meV, resulting in $J_F/J_0 \approx 1.5$. $t \approx 1$ eV

is the transfer matrix element between neighbouring copper and oxygen sites, $U \approx 8\,\mathrm{eV}$ is the Coulomb repulsion for two holes at the same copper site (Hubbard repulsion), and $\epsilon = \epsilon_p - \epsilon_d \approx 3\,\mathrm{eV}$ is the difference between the on-site energies on oxygen and copper sites in the way it enters the three-band Hubbard model.

1.1. Spin wave picture

An unperturbed antiferromagnet is usually described by introducing two sublattices, a bosonization of the spin excitation operators (S^+, S^-) via the Holstein–Primakov approximation and the neglection of two-particle interactions. This leads to the Hamiltonian

$$\begin{aligned} H &= E_0 + \epsilon_1 \sum_i a_i^+ a_i + \epsilon_2 \sum_j b_j^+ b_j + \\ &+ J \sum_{\langle i,j \rangle} (a_i^+ b_j^+ + h.c.) \end{aligned} \tag{4}$$

with $\epsilon_{1/2} = g\mu_B(H_A \pm H_0) + JzS^2$, the anisotropy field H_A confirming that the Neel state is the ground one, $a_i^+, b_j^+ (a_i, b_j)$ being the creation (annihilation) operators for bosons.

This Hamiltonian is usually solved in k space with the Bogolyubov transformation leading to the well-known AF magnon bands with a dispersion relation $\epsilon(k) \sim |\sin(ka)|$.

1.2. Local perturbation

In order to introduce the local perturbation that results from turning a particular spin and switching on a ferromagnetic exchange interaction around it, we follow the ideas of Lovesey [9]. The turning of a spin means subtracting this spin from one sublattice and adding it to the other. Therefore, the turning of a spin at site $_0$ leads to the perturbation Hamiltonian at this site:

$$\begin{aligned} H_p &= E_{p0} + \Delta\epsilon_1 a_0^+ a_0 + \Delta\epsilon_2 \sum_{\langle 0,j \rangle} b_j^+ b_j - \\ &- J \sum_{\langle 0,j \rangle} (a_0^+ b_j^+ + h.c.) - \\ &- J_F \sum_{\langle 0,j \rangle} (a_0^+ b_j + h.c.). \end{aligned} \tag{5}$$

Here $\Delta\epsilon_1 = -2g\mu_B H_0 - 2J_F - 2J$ and $\Delta\epsilon_2 = -J_F - J$ describe the changes of diagonal energies.

1.3. Green's functions

Since we want to calculate the local Green's functions, G_{ij}, we rewrite the equation of motion $(\omega \mathbf{I} - \mathbf{H})\mathbf{G} = \mathbf{I}$ as it was suggested in [10]. Therefore we form 2x2 packages of the local Green's functions:

$$P_{ij} = \begin{pmatrix} \langle\langle \alpha_i^+, \beta_j \rangle\rangle & \langle\langle \alpha_i^+, \beta_j^+ \rangle\rangle \\ \langle\langle \alpha_i, \beta_j \rangle\rangle & \langle\langle \alpha_i, \beta_j^+ \rangle\rangle \end{pmatrix}, \quad \alpha, \beta \in \{a, b\}. \tag{6}$$

The part where the Hamiltonian enters the equation of motion gets the following form:

$$D_{ij}^{AF} = \begin{pmatrix} & J^{AF} \\ -J^{AF} & \end{pmatrix} \quad \text{for an antiferromagnetic,}$$

$$D_{ij}^{F} = \begin{pmatrix} J^{F} & \\ & -J^{F} \end{pmatrix} \quad \text{for a ferromagnetic} \tag{7}$$

interaction between the sites i and j, while

$$D_{ii}^{D} = \begin{pmatrix} -\epsilon_k & \\ & \epsilon_k \end{pmatrix}, \quad k \in \{1, 2\}, \tag{8}$$

is the diagonal part of D.

In this way the equation of motion of the Green's function reads:

$$\sum_j \left[\begin{pmatrix} \omega & \\ & -\omega \end{pmatrix} \delta_{ij} - D_{ij}^{\lambda} \right] P_{ij} = \delta_{ij}, \tag{9}$$

$$\lambda \in \{AF, F, D\}.$$

1.4. Unperturbed Green's functions

For a two-dimensional case the unperturbed local Green's functions are given by

$$G_{ii}^{-+a} = \langle\langle a_i a_i^+ \rangle\rangle = \frac{4}{\pi^2} \int_0^{\pi} \int_0^{\pi} dk \frac{D_2}{D_2 D_1' + J(k)^2}, \tag{10}$$

$$G_{jj}^{-+b} = \langle\langle a_j a_j^+ \rangle\rangle = \frac{4}{\pi^2} \int_0^{\pi} \int_0^{\pi} dk \frac{D_1}{D_1 D_2' + J(k)^2}, \tag{11}$$

$$D_{1/2} = \frac{\omega + \epsilon_{1/2}}{J}, \qquad D_{1/2}' = \frac{\omega - \epsilon_{1/2}}{J},$$

$$J(k) = 4 \cos k_x a \cos k_y a,$$

which results in the elliptical integrals:

$$G_{ii}^{-+a} = \frac{2D_2}{\pi\sqrt{D_2D_1'(1-D_2D_1')}}\mathbf{IK}\left(\frac{1}{\sqrt{1+D_2D_1'}}\right), \tag{12}$$

$$D_2D_1' > 0.$$

$$G_{jj}^{-+b} = \frac{2D_1}{\pi\sqrt{D_1D_2'(1-D_2D_2')}}\mathbf{IK}\left(\frac{1}{\sqrt{1+D_1D_2'}}\right), \tag{13}$$

$$D_1D_2' > 0.$$

Further Green's functions follow from the equation of motion.

1.5. Dyson equation

The perturbation formed by a single turned spin with four local ferromagnetic interactions around it as sketched in Fig. 1 is given by a 10-dimensional matrix:

$$G^0 = G(1 - H^S G^0), \tag{14}$$

$$P_{ij}^0 = \sum_k P_{ik}\left(\begin{pmatrix} \omega & \\ & -\omega \end{pmatrix}\delta_{kj} - \sum_l \Delta D_{kl} P_{lj}^0\right), \tag{15}$$

where ΔD_{kl} is given by

$$\Delta D_{kk} = \begin{pmatrix} -\Delta\epsilon_{1/2} & \\ & \Delta\epsilon_{1/2} \end{pmatrix}, \quad \epsilon_1 \text{ for } k = 0, \epsilon_2 \text{ otherwise;}$$

$$\Delta D_{kl} = \begin{pmatrix} J^F & -J^{AF} \\ J^{AF} & -J^F \end{pmatrix},$$

$$\text{for } k = 0, l \in \{1,2,3,4\}$$

$$\text{or } l = 0, k \in \{1,2,3,4\};$$

$$\Delta D_{kl} = \begin{pmatrix} 0 & 0 \\ 0 & 0 \end{pmatrix} \quad \text{otherwise.}$$

1.6. Localized states

The zeros of the determinant

$$\mathrm{Det}\left|\begin{pmatrix} \omega & \\ & -\omega \end{pmatrix}\delta_{kj} - \sum_l \Delta D_{kl} P_{lj}^0\right| \tag{16}$$

give localized dispersionless states that split from AF magnon bands, as is shown in Fig. 3. For the values of $J_F/J_0 \approx 1.5$, especially above the magnon band, such states result from this calculation. Experimentally these states might be observable by neutron scattering, but this may be difficult since at the low doping concentration regarded in the calculation there will be an extremely low intensity of the measured signal.

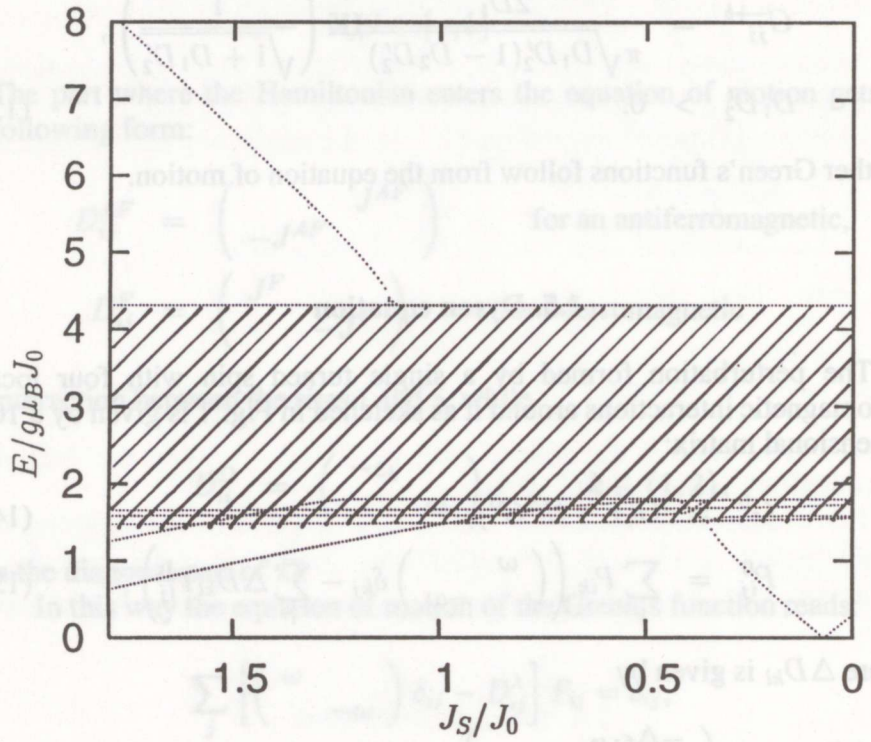

Fig. 3. Localized dispersionless states that split up from the AF magnon band (hatched region) for an external magnetic field of $H_0/J_0 = 0.01$ and an anisotropic field $H_A/J_0 = 0.3$. The figure is drawn about different values of the magnetic impurity exchange coupling, J_S. The relevant part is the region for $J_S/J_0 \geq 1.5$ (see the text).

1.7. Magnetic susceptibility

We calculate the influence of such ferronic perturbation on magnetic susceptibility. The magnetization of the system is given by

$$M(H_0, T) = \frac{1}{2} \sum_i g\mu_B \left(1 - \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{1}{e^{\beta\omega} - 1} \mathrm{Im} G^{ii}(\omega, H_0) d\omega\right), \quad (17)$$

while susceptibility is

$$\chi(T) = \frac{\partial M(H_0, T)}{\partial H_0}|_{H \to 0}. \quad (18)$$

Since $G^{ii}(\omega, H_0)$ is the only part in Eq. (17) that contains the external magnetic field H_0, we emphasize the expression

$$\Delta G^{ii} := \lim_{H_0 \to 0} \frac{G^{ii}(H_0) - G^{ii}(0)}{H_0}. \tag{19}$$

Neglecting all terms of higher order in H_0, ΔG^{ii} turns out as a linear combination of elliptical integrals, resulting for ΔG^{00} for example in:

$$\begin{aligned}(\Delta G^{00})^0 \;=\; & \frac{4}{\pi} \frac{16J^2}{(\omega^2 - \epsilon^2)\sqrt{p(1-p)}} \times \\ & \times \left[\left((\omega + \epsilon)^2 - (\omega^2 - \epsilon^2) \right) \mathbf{IK}\left(\sqrt{\frac{1}{1-p}} \right) + \right. \\ & \left. + \left((\omega + \epsilon)^2 + (\omega^2 - \epsilon^2) \right) \mathbf{IE}\left(\sqrt{\frac{1}{1-p}} \right) \right]. \end{aligned} \tag{20}$$

Here $\epsilon = \epsilon_1(H_0 = 0) = \epsilon_2(H_0 = 0)$ and $p = -\frac{\omega^2 - \epsilon^2}{16J^2} < 0$. For other values of p the elliptical integrals are continued over the complex plane. The additional part of susceptibility reveals weak temperature dependence and is opposite to the part from the AF magnons, whereby it decreases with the increase of temperature (see Fig. 4).

1.8. Total magnetic susceptibility

We add up the terms of magnetic susceptibility and add the Curie-type contribution of the weak interacting magnetic impurities

$$\chi_{\text{imp}}(T) \approx \frac{n(Sg\mu_B)^2}{kT}. \tag{21}$$

The unperturbed part, $\Delta G^{ii\,0}$, gives the increase of susceptibility towards the Neel temperature (see Fig. 4). The perturbed part of ΔG^{ij} is now again obtained by the Dyson equation and reads as follows:

$$\Delta G^{ij} \approx \Delta G^{ij\,0} \left(\underline{\underline{1}} - H^S G^{ij\,0} \right)^{-1}, \tag{22}$$

with $n = \delta$ the concentration of impurities, $S = 3/2$ (or 2), depending on the model of the magnetic polaron used (see e.g. [11]). The result can be seen in Fig. 5, that maps qualitatively the experimental observations of Fig. 2.

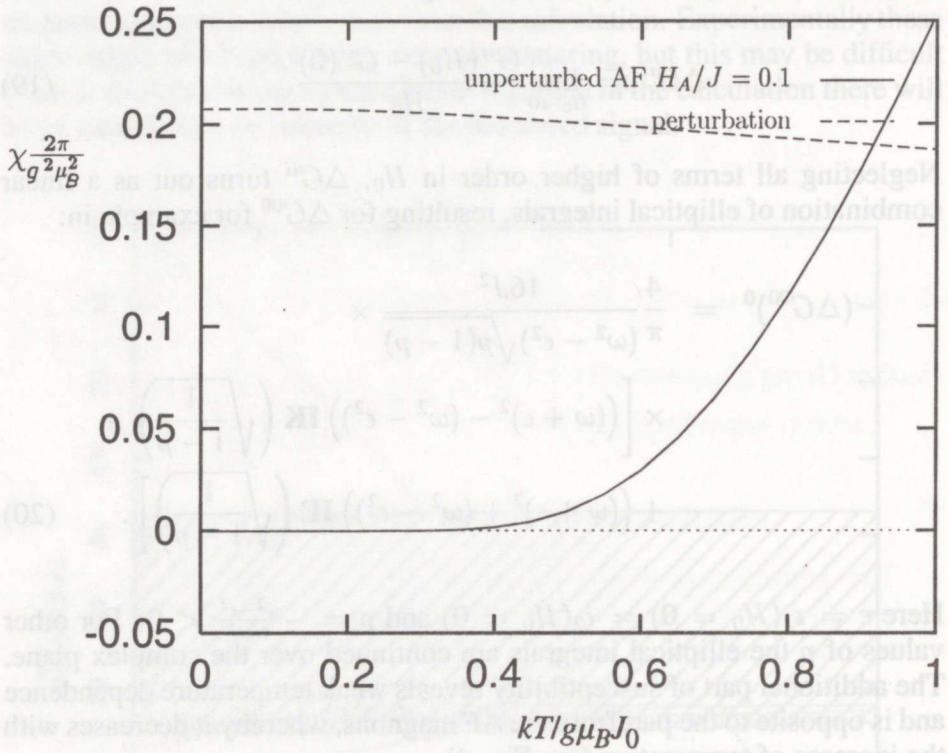

Fig. 4. The calculated magnetic susceptibility for the uperturbed AF as a usual quadratic increase towards the Neel temperature. The perturbed part (dotted line) is the average of nine lattice sites around perturbation.

CONCLUSION

The model of percolative phase separation is able to explain the temperature and doping dependence of magnetic susceptibility. The susceptibility consists of three parts. One part decreasing with doping comes from the AF magnons and leads to the increase of susceptibility to the Neel temperature. The second, the Curie part (χ_{imp}), is given by weak interacting magnetic polarons. The third, the intermediate range, is due to the perturbation of the AF magnon spectrum.

From this picture the creation of dispersionless localized states occurring above the AF magnon spectrum in a very low doping regime is predicted. These localized states should be observable with, e.g. neutron scattering, although the expected intensity will be very small due to low doping.

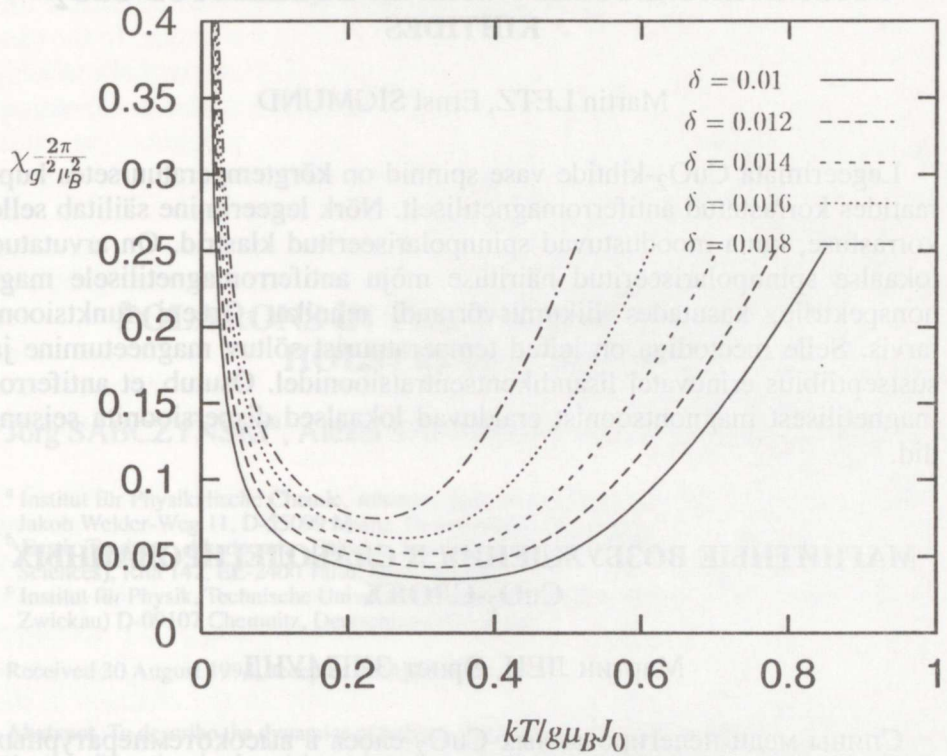

Fig. 5. Total calculated magnetic susceptibility for different doping concentrations. Note that with the exception of some temperature-independent background susceptibility the exact lineshape of the experimental data (see Fig. 2) was obtained.

ACKNOWLEDGEMENT

The authors wish to thank M. V. Eremin from Kazan State University for fruitful discussions.

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MAGNETERGASTUSED NÕRGALT LEGEERITUD CuO_2 - KIHTIDES

Martin LETZ, Ernst SIGMUND

Legeerimata CuO_2-kihtide vase spinnid on kõrgtemperatuursetes kupraatides korrastatud antiferromagnetiliselt. Nõrk legeerimine säilitab selle korrastuse, kuna moodustuvad spinnpolariseeritud klastrid. On arvutatud lokaalse spinnpolariseeritud häirituse mõju antiferromagnetilisele magnonspektrile, kasutades liikumisvõrrandi tehnikat Greeni funktsiooni tarvis. Selle meetodiga on leitud temperatuurist sõltuv magneetumine ja sustseptiiblus erinevatel lisandikontsentratsioonidel. Osutub, et antiferromagnetilisest magnontsoonist eralduvad lokaalsed dispersioonita seisundid.

МАГНИТНЫЕ ВОЗБУЖДЕНИЯ В СЛАБОЛЕГИРОВАННЫХ CuO_2-СЛОЯХ

Мартин ЛЕЦ, Эрнст ЗИГМУНД

Спины меди нелегированных CuO_2-слоев в высокотемпературных купратах упорядочены антиферромагнитно (АФ). Слабое легирование сохраняет эту упорядоченность, так как образуются спин-поляризованные кластеры. Вычислено влияние локального спин-поляризованного возмущения на АФ-магнонный спектр, используя технику уравнения движения для функции Грина. Этим методом найдены температурно-зависимые намагничивание и восприимчивость при разных концентрациях примеси. Оказывается, что от АФ-магнонных зон расщепляются локальные бездисперсионные состояния.