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INELASTIC NEUTRON SCATTERING
AND ANTIFERROMAGNETIC FLUCTUATIONS
IN HIGH TEMPERATURE SUPERCONDUCTORS

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1 Introduction

In the present paper the results are discussed of the neutron scattering experiments on investigation of antiferromagnetic (AF) fluctuations. In the last few years these have been exciting intense interest because of the following.

The common feature of high temperature superconductors is the antiferromagnetic ordering of spins of Cu ions in CuO_2 planes (Fig.1). In the dielectric phase the long-range order takes place because of both the presence of spin $S = 1/2$ of a hole in the 3d-shell of copper in the Cu^{2+} state and the strong indirect (via oxygen ions) exchange interaction. In the metallic and the superconducting phase the long-range magnetic order disappears, though very strong dynamic spin fluctuations remain in a rather wide range of temperatures. This fact gives rise to models of superconducting pairing via magnetic degrees of freedom.

The most complete information about magnetic structure and spin correlations is obtained in neutron scattering experiments. The investigations of lanthanum and yttrium compounds were most detailed. Large single crystals were grown, which is of great significance to investigation of dynamical phenomena. Recently a number of inelastic neutron scattering data have appeared in confirmation of the special role of AF fluctuations in these superconductors.

2 Inelastic Neutron Scattering Study of Magnetic Excitations

In experiments the spin waves near the antiferromagnetic state at $x < 0.4$ and the spectra of AF fluctuations in the metallic phase at various concentrations of holes, including the superconducting phase were measured. Most exciting was the observation[1] of a gap in the spectrum of spin fluctuations below the temperature of the superconducting transition, T_c . These were carried out by the French group headed by Prof. J.Rossat-Mignod, the eminent physicist and director of LLB (Saclay), whose tragic death in the August of this year is a great loss to all of us.

Figure 2 shows, for different temperatures, the energy dependence of the imaginary part of the dynamical spin susceptibility, $\text{Im}\chi(Q, \omega)$ [2], for the superconductor $\text{YBa}_2\text{Cu}_3\text{O}_{6.69}$ with $T_c = 59\text{K}$. It is seen that at low energies the intensity of fluctuations strongly decreases, which can be interpreted as being due to the appearance of a gap in the spectrum at $T < T_c$. The insert to the figure gives a clear picture of this. The gap width $E_G \approx 1 \div 3kT_c$ is comparable with, though smaller than, that of the superconducting gap in the plane $2\Delta_{ab} \approx 6kT_c$.

The fact of the existence of a gap in the spectrum of magnetic excitations is confirmed by independent measurements of the Brookhaven group[3], including those carried out at $x = 0.92$ and $T_c = 91\text{K}$. Recently a paper was published[4] reporting the results of analogous measurements performed at Oak Ridge, but with polarized neutrons, which, of course, better answers the needs of the problem. The results reported in[4], though different from those of work[3], are also evidence

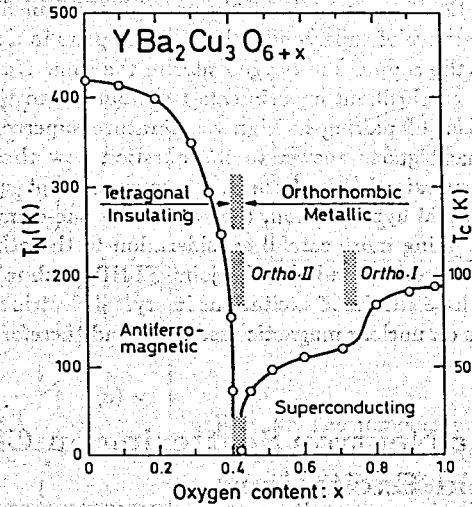


Fig. 1. The phase diagram of the $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ system as a function of oxygen content x (from ref.[1]).

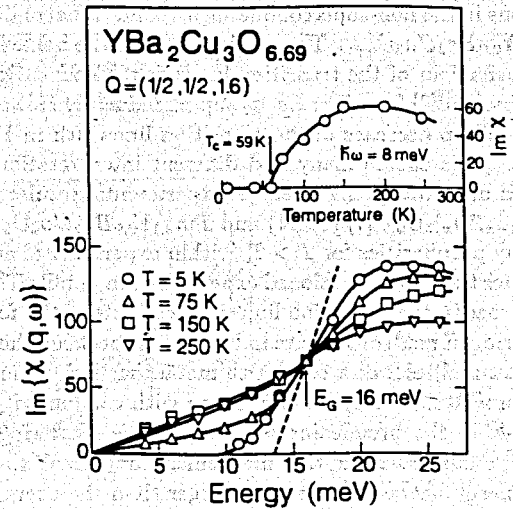
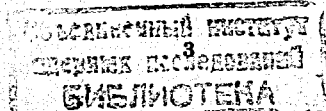


Fig.2. The low energy part of the spin fluctuation spectrum for $\text{YBa}_2\text{Cu}_3\text{O}_{6.69}$ (from ref.[2]).



for certain connections between the antiferromagnetic spin fluctuations of $Cu2$ ions and the appearance of superconducting Cooper pairs in a strongly correlated system of holes in the copper and oxygen sites in the same CuO_2 plane.

Unfortunately, the brilliant experiments[1-4] pointing to the existence of the magnetic mechanism of pairing in high temperature superconductors have, up to now, not an unambiguous answer to the question how these two subsystems interrelate: do they exist in the form of a two-component spin liquid or, having very strong mutual hybridization, they exist as a one-component spin liquid. Therefore, we are giving more careful consideration to the other experiments on inelastic neutron scattering made by the joint FLNP (Dubna) — RAL (Didcot, UK) group[5-8]. These studies of excitations in crystal electric fields are very close to the experiments on nuclear magnetic resonance and therefore can be compared with them.

3 Inelastic Neutron Scattering on Crystal Electric Field Excitations

Inelastic neutron scattering on crystal field (CF) excitations in Tm substituted $YBa_2Cu_3O_{7-x}$ attracts attention of scientists because of the strange behaviour of the transition linewidth in dependence on temperature [5,6] as shown in Fig.3.

This figure illustrates the results of measurements of the transition linewidth for the excitations in the non-superconducting (a) and superconducting (b) state of a sample $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6+x}$. The solid line marks the behaviour similar to the corresponding behaviour of the transition linewidth in conventional superconductors, like that observed[9] for a $Tb : LaAl_2$ superconductor. However, qualitatively, they differ by a sharp decrease of the transition linewidth in $YBCO$, which occurs at $T > T_c$. This causes many and different interpretations. The situation has become even more intriguing after the experimental results were obtained for $Tm_{0.1}Y_{0.9}Ba_2(Cu_{1-x}Zn_x)_3O_{6.9}$ [7] (Fig.4) and $Tm_{0.1}Y_{0.9}Ba_2Cu_4O_8$ [8] (Fig.5) which did not show any peculiarities for $T > T_c$ within experimental error.

Let us consider the above mentioned experiments in detail. The physical reason for a sharp decrease in the transition linewidth consists in the following. Inelastic magnetic scattering of neutrons occurs in transitions between the crystal field split levels of the main multiplet of a rare-earth metal ion. Relaxation of the localized $4f$ magnetic moment caused by its interaction with conductivity electrons ($s-f$ interaction) leads to the broadening of peaks in the scattering spectra. In the superconducting state this relaxation mechanism may break the Cooper pairs, if the transition energy between levels, ϵ , is larger than the energy gap, $2\Delta(T)$. In this case the appearance of a gap in the spectrum will not affect the temperature dependence of the transition linewidth.

When $\epsilon < 2\Delta(T)$, the excitation energy, ϵ , is insufficient to break the Cooper pairs. As a result the relaxation channel via the $s-f$ interaction is switched off, which fact leads to a sharp decrease in the transition linewidth of the cor-

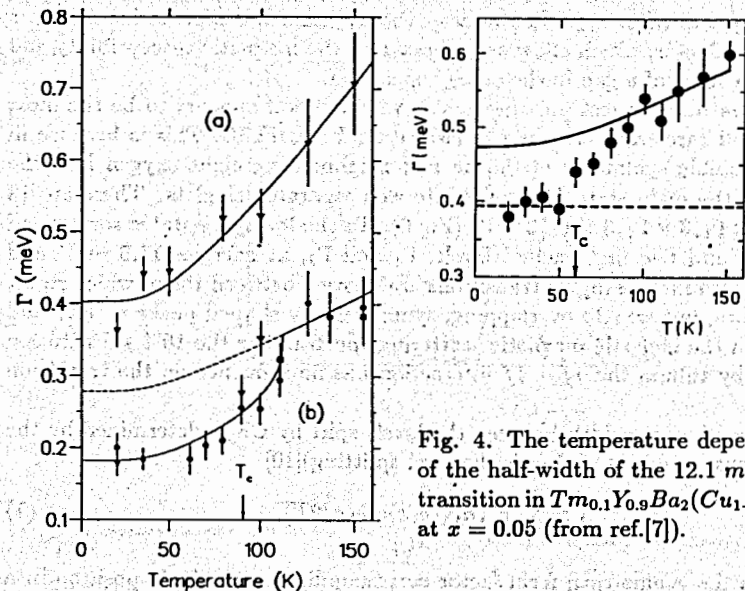


Fig. 3. The temperature dependence of the half-width Γ (a) of the 13.8 meV CF transition in $Tm : YBa_2Cu_3O_{6.1}$ and (b) of the 11.8 meV CF transition in $Tm : YBa_2Cu_3O_{6.9}$ (from ref.[6]).

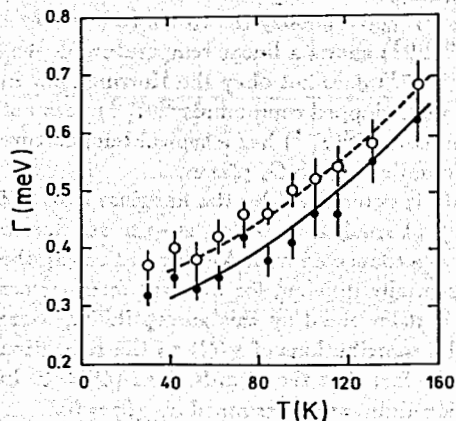


Fig. 4. The temperature dependence of the half-width of the 12.1 meV CF transition in $Tm_{0.1}Y_{0.9}Ba_2(Cu_{1-x}Zn_x)_3O_{6.9}$ at $x = 0.05$ (from ref.[7]).



Fig.5. The temperature dependence of the linewidth in $YBa_2Cu_4O_8$ (the lines show the results of the model calculations: the dashed line for $\epsilon = 11.8 \text{ meV}$, the solid line for $\epsilon = 14.3 \text{ meV}$) (the experimental points from ref.[8]).

responding peaks below T_c . In this way the $4f$ moments are the local probes in a superconductor, which allow one to measure the magnetic susceptibility and estimate the value of a gap in the energy spectrum.

It appears that for test measurements the Tm^{3+} ion appears to be the most suitable of all rare-earth elements to substitute Y in HTSC. This is because in the orthorhombic symmetry of the nearest neighbouring eight oxygen ions the multiplet of the main state 3H_6 splits into well separated singlets. There are 13 singlets: $4 \times \Gamma_1, 3 \times \Gamma_2, 3 \times \Gamma_3, 3 \times \Gamma_4$ (Fig.6). To the low-lying states correspond the level Γ_3 and two first excited levels, Γ_4 and Γ_2 , at energies 11.8 meV and 14.2 meV. Two intense dipole transitions are allowed between them, which result in two intense, but weakly overlapping, symmetrically shaped peaks at 11.8 and 14.2 meV in the inelastic magnetic scattering spectra. For the 10% substitution of yttrium by tulium the $4f-4f$ interaction has no influence on the transition linewidth.

The transition linewidth between the levels split by CF is determined by the dynamical susceptibility $\chi(\epsilon)$ (ϵ is the level splitting)[10]

$$\gamma \propto \sum_q F_q \text{Im} \chi(q, \epsilon) \text{cth}(\epsilon/2T), \quad (1)$$

where F_q is the geometrical form factor corresponding to the ionic position in a unit cell.

In the case of $YBaCuO$, eq.(1) describes the linewidth of CF transition caused by both the interaction with an excitation in a subsystem of local spins of Cu ions in CuO_2 planes, and the interaction with conducting p -holes.

Note that for $\omega \rightarrow 0$ the spin lattice relaxation rate T_1^{-1} is proportional to the dynamical susceptibility also, but that means that for a small splitting ϵ of the $4f$ levels, the linewidth of the CF transition and the spin-lattice relaxation rate should show similar temperature dependences.

The spin-lattice relaxation rates for ^{89}Y and ^{17}O obey the Korringa law over a wide enough temperature region[11]. $^{89}(T_1^{-1})$ shows a linear temperature dependence in nonmetallic compounds[12]. $^{63}(T_1^{-1})$ does not obey the Korringa law in any temperature region. Note that for underdoped compounds $^{63}(T_1^{-1})$ does not exhibit any peculiarities at $T = T_c$. Moreover, $^{63}(T_1^{-1})$ has a typical temperature $T^* \sim 150K$ [11], connected with AF fluctuations in CuO_2 planes.

Since the spin-lattice relaxation rate is determined by the imaginary part of the dynamical susceptibility, $\text{Im} \chi(q, \omega)$, at small frequencies, $\omega \rightarrow 0$, so, due to large increases of χ for large wavevectors q connected with AF fluctuations[2], the spin-lattice relaxation rates can be essentially different for different form-factors $F(q)$. According to[11,13,14] $^{63}(T_1^{-1})$ is determined by this susceptibility, when the AF wavevector $Q = (\pi, \pi)$, and the contributions of $\chi(Q)$ to the relaxation rates of ^{17}O and ^{89}Y are small due to the fact that the form-factor $F(Q) = 0$. In this latter case the spin-lattice relaxation times are determined by $\chi(q = 0)$.

To better understand the temperature dependence of relaxation rates and $4f$ transition widths the temperature dependence of the static susceptibility and the

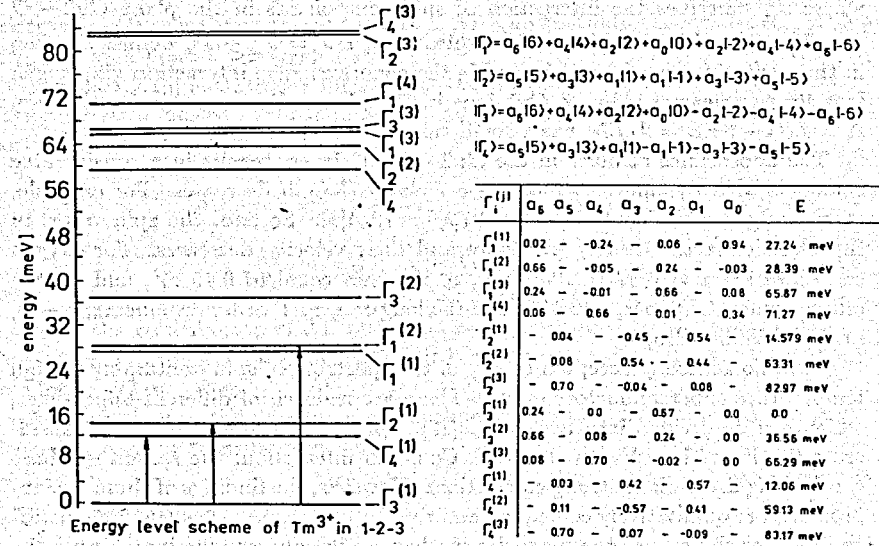
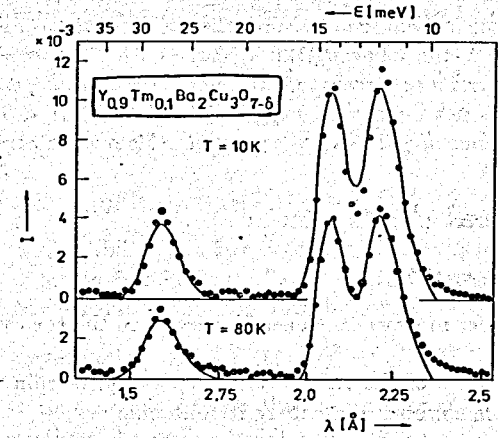


Fig. 6. The energy level scheme of Tm^{3+} in $YBa_2Cu_3O_{7-\delta}$ and the dependence of the scattering intensity on the neutron wave length λ .

Knight shift need to be known. The main findings of the susceptibility and Knight shift measurements are the following[12]: i) ΔK is temperature dependent and this temperature dependence corresponds to the temperature dependence of χ ; ii) the ratio $\Delta K/\chi$ does not depend on oxygen content for $6.41 < x < 7$; iii) the temperature dependence of $\chi(T)$ changes slowly in this oxygen content region.

4 Theoretical Analysis

4.1 General Remarks

First, let us consider in short the general situation in the theory of AF fluctuations in HTSC.

In the dielectric state of the AF phase the spectrum of spin waves may be described with the anisotropic Heisenberg Hamiltonian

$$H = - \sum_{ij} J_{ij}^{\parallel} S_i^{\parallel} \cdot S_j^{\parallel} + J_{ij}^{\perp} (S_i^x S_j^x + S_i^y S_j^y) / 2 \quad (2)$$

where J_{ij}^{\perp} describes the interaction of spin components in the plane $Cu2 - O$ and J_{ij}^{\parallel} — outside this plane. The anisotropy, $\Delta J = J^{\perp} - J^{\parallel}$, defines the gap in the excitation spectrum, where J is the superexchange interaction via oxygen ions for two nearest spins of $Cu2$ ions in the plane. Correspondingly, the gap, $\Delta = J\sqrt{1 - \alpha^2}$, $\alpha = J_{\perp}/J_{\parallel}$, has a small value: $\Delta < 0.5 \text{ meV}$.

The appearance of holes in the $Cu2 - O_{2,03}$ layers results in a considerable change of spin dynamics. First, the correlation length decreases. For example, at an oxygen concentration $x = 6.37$, $\xi = 7.5 \text{ \AA}[1]$. Second, the spin waves in the plane start to strongly attenuate and their velocity decreases. For oxygen concentration $x = 6.37$ this velocity, v , becomes equal to 0.45 eV , and for the critical concentration $x_c = 6.41$, when the long-range AF order disappears, $v \rightarrow 0$, i.e. spin rigidity drops.

Now, no generally accepted theory of the spectrum of spin excitations in high temperature superconductors exists. There are a dozen of different approaches, which can be divided into two groups[15]. The first includes the works considering the Fermi-liquid with the weak Coulomb interaction, the second — those considering the limit of strong correlations. However, the findings of these two approaches are qualitatively coinciding and result in the appearance of a "magnetic" gap. As for the role of the magnetic mechanism in superconducting pairing, the theory in its present state is far from being able to give a quantitative criterium for comparison of its predictions with experiment. At the same time it has been established that the indirect interaction of charge carriers via AF spin fluctuations could cause the pairing. The most probable exhibition of the magnetic mechanism is the d -pairing, though, under certain assumptions, it might also give rise to the attraction via the s -channel.

So, at present, it seems of interest to clear up the qualitative basis of the state of the spin system in HTSC. Rather promising here is the phenomenological theory

of the AF Fermi-liquid[14]. The authors of[14] suggested the introduction of model dynamical spin susceptibility in the form:

$$\chi(q, \omega) = \chi_{QP}(q, \omega) + \chi_{AF}(q, \omega), \quad (3)$$

where χ_{QP} stands for the quasiparticle contribution

$$\chi_{QP}(q, \omega) = \chi_0(T) [1 - i\omega\pi/\Gamma]^{-1} \quad (4)$$

$\chi_0(T)$ is the static susceptibility, and Γ/π is the typical energy of spin fluctuations, which value is close to the Fermi energy, E_F .

The susceptibility, χ_{AF} , describes the contribution of AF fluctuations

$$\chi_{AF}(q, \omega) = \chi_Q [1 + \xi^2(Q - q)^2 - i\omega/\omega_{SF}]^{-1}, \quad (5)$$

where χ_Q is the static susceptibility for the wave vector, Q , $\hbar\omega_{SF}$ is the typical energy of AF fluctuations. According to[14] a relation

$$\chi_Q = \chi_0(T)(\xi/\xi_0)^2, \omega_{SF} = (\Gamma/\pi)(\xi_0/\xi)^2 \quad (6)$$

exists, where $\xi(T)$ is the correlation length of AF spin fluctuations, because $(\xi/\xi_0)^2 \gg 1$, $\chi_Q \gg \chi_0(T)$ and $\Gamma \gg \omega_{SF}$.

The theory[14] successfully interpreted the results of the NMR experiments, particularly the behaviour of the spin-lattice relaxation rate, T_1^{-1} , which is proportional to the function

$$S(q, \omega \rightarrow 0) = \lim_{\omega \rightarrow 0} \frac{kT}{\hbar\omega} \text{Im} \chi(q, \omega + i\epsilon) = \pi \frac{kT\chi_0(T)}{\hbar\Gamma(T)} \left[1 + \frac{\beta(\xi(T)/a)^4}{1 - \xi^2(T)(Q - q)^2} \right] \quad (7)$$

In eq.(7) the contribution of AF fluctuations is determined by the second term, proportional to the weight coefficient, β . It is small for $q = 0$, and plays a decisive role for $q = Q_{AF}$.

Now, we turn to the measurements outlined in Section 3 for the temperature dependence of the transition linewidth.

4.2 The Dielectric Phase

The compound $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.1}$ is an AF insulator and the main contribution to the broadening of transitions in CF arises from magnetic fluctuations. To describe the $4f$ transitions we introduce the Hamiltonian similar to ref.[13]:

$$H = D \sum_{k=1}^8 S_k \sigma, \quad (8)$$

where S_k is the spin operator of a Cu ion, σ are the Pauli matrices, describing the transition between the $4f$ levels split by CF, and D is the interaction constant.

Summation is over the eight nearest neighbouring Cu ions. We also adopt the anisotropic Heisenberg Hamiltonian (2) for describing the spin dynamics in CuO_2 .

In the pure AF state the gap is sufficiently small $\Delta < 5 \text{ meV}$. In this case the temperature dependent contribution to the linewidth is determined by the processes of two types. For $\Delta > \epsilon$ the main contribution arises from the Compton processes which describe the linewidth in the superconducting state. Note that according to [13] the spin-lattice relaxation rate can be described by the Compton processes also. For $\Delta < \epsilon$ the main contribution arises from the one-magnon emission-absorption processes. On the other hand the spin-lattice relaxation rate is also determined by the Compton processes. This means that these two values have different temperature dependences in the AF state

$$\gamma_{\Delta < \epsilon} = 2\pi D^2 f(\epsilon) (2n(\epsilon) + 1) |u(\epsilon) - v(\epsilon)|^2, \quad (9)$$

where $f(\epsilon) = 8 \int dq^2 / (2\pi)^2 \delta(\omega - \epsilon(q)) (1 + \cos(q_x) + \cos(q_y) + \cos(q_x)\cos(q_y))$, $u(\omega) = [\frac{1+\omega}{2\omega}]^{1/2}$, $v(\omega) = [\frac{1-\omega}{2\omega}]^{1/2}$, $n(\omega) = 1/(exp(\omega/T) - 1)$.

Calculations of the linewidth using eq.(9) (see Fig.7) show that one magnon processes describe the experimental value of the linewidth in $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.1}$ for all temperatures. The inhomogeneous broadening is $\gamma_0 = 0.17 \text{ meV}$ and $D = 0.03J \sim 4 \text{ meV}$. Moreover, the best agreement with experiment requires a small value for the spin gap, $\Delta < 5 \text{ meV}$. That value is in good agreement with the experimental data reported in [2].

Therefore in the AF state the CF transition linewidth is described within the spin-wave approximation and the main contribution arises from one magnon processes. This means that the spin gap is small compared to the $4f$ level splitting.

4.3 The Superconducting Phase

To analyze the temperature dependence of the linewidth in superconducting compounds we use the phenomenological approach [14] with the approximation for $\chi(q, \omega)$ as given in Eq.(7).

The temperature dependence of the correlation length in (7) has the form:

$$\left(\frac{\xi(T)}{a}\right)^2 = \left(\frac{\xi_0}{a}\right)^2 \left(\frac{|T_x|}{T + T_x}\right). \quad (10)$$

Note that the investigation of AF correlation in superconducting materials [2] does not provide for such type of dependence. However, for the analysis of the temperature dependence of the $4f$ transition linewidth, the contribution of $\chi(Q)$ is suppressed due to the fact that $F(Q) = 0$ and the temperature dependence of the correlation length is not essential.

Also note that according to ref.[11] the squared Lorentzian proposed in [14] for the Q dependence of the susceptibility (7) is definitely too wide to explain the temperature dependence of $1/T_1 T^{-1}$. According to ref.[3] the Q dependence of susceptibility is described by a Gaussian and falls off much faster than a Lorentzian

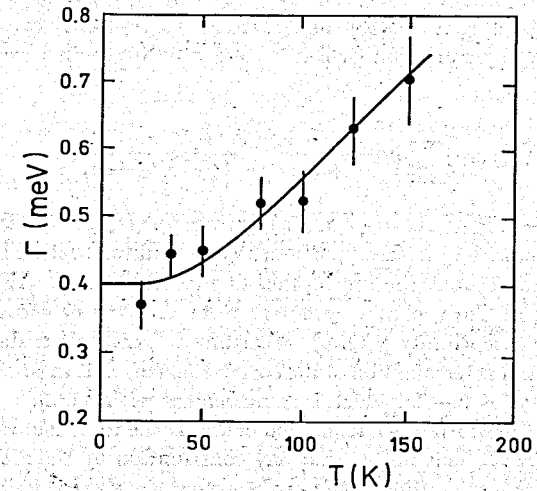


Fig. 7. The temperature dependence of the linewidth for the insulating state $x = 6.1$. The experimental points from ref.[6]. The solid line shows the result of the model calculations.

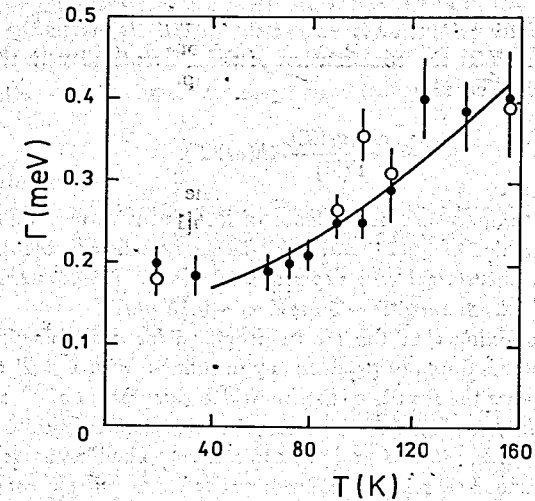


Fig. 8. The temperature dependence of the linewidth for the superconducting state $x = 6.9$. The experimental points from ref.[6]. The solid line shows the result of the model calculations.

Q dependence. According to ref.[11] the dynamical susceptibility can be approximated by the following expression (which is supported by the neutron scattering experiments[3]):

$$\chi(q) = \frac{\pi}{4} \sum_{i=1}^4 \frac{\chi_0(T) + \chi(Q^i)\phi(Q^i - q)}{\Gamma(T) + (\Gamma(T) - \Gamma_{SF})\phi(Q^i - q)}, \quad (11)$$

where $\phi(q) = \exp(-\ln 2 \xi^2 q^2)$, $Q^i = (\pi \pm \delta, \pi)$, $(\pi, \pi \pm \delta)$, $\chi(Q^i)$ is the susceptibility for the AF wavevector, Γ_{SF} is the typical energy of spin fluctuation for Q^i . Note that (10) takes into account the appearance of incommensurate AF fluctuation under the doping of CuO_2 planes by holes. Note that the Q width of the AF Gaussian peak of susceptibility $\chi(Q, \omega)$, according to ref.[3], is ω dependent, so that a straightforward determination of the correlation length is impossible. Rough estimates show that $\xi \sim 2 - 3a$, which is in agreement with ref.[2].

Eq. (1) for the linewidth of $4f$ transition contains the form factor $F(q)$ which is equal to 0 for $q = Q$. This means that any contribution by the second term of (11) is small. According to estimates of the absolute value of spin lattice relaxation rates for $^{63}(T_1^{-1})$ and $^{17}(T_1^{-1})$ the contribution is less than 0.01[11]. Indeed, the contribution of the second term in eq.(11) can be easily estimated as:

$$1/(16\pi(\ln 2 \xi^2)^3) \cdot \chi(Q_{AF})/\Gamma(T). \quad (12)$$

The numerical value for $\xi \sim 2 \div 3a$ is smaller than 0.001. It means that due to the filtering form factor $F(Q)$ the contribution of the susceptibility to the AF wave vector is strongly suppressed

$$\gamma \sim \frac{\epsilon D^2 \chi_0(T)}{\Gamma(T)} \text{cth}(\epsilon/2T). \quad (13)$$

To analyze the temperature dependence of linewidth we use eq.(13) and the temperature dependence of $\chi_0(T)$ and $\Gamma(T)$ as presented in ref.[14]. The best agreement between theoretical and experimental results (Fig.8) corresponds to the following set of parameters: $D \sim 5 \text{ meV}$, $\gamma_0 \sim 0.15 \text{ meV}$.

It is worth mentioning that the model proposed for describing linewidth in high- T_c superconductors does not produce any peculiarities at $T > T_c$ as discussed in [5,6]. Figure 8 shows the results of the linewidth calculation of $4f$ transition in Tm substituted $YBa_2Cu_4O_8$, in comparison with the experimental data[8]. The temperature dependence, $\chi(T)$, is presented in ref.[16]. The best agreement of the theoretical and experimental results corresponds to the following set of parameters: $D \sim 6 \text{ meV}$, $\gamma_0 = 0.27 \text{ meV}$ for the $\epsilon = 11.8 \text{ meV}$ transition and $\gamma_0 = 0.31 \text{ meV}$ for the $\epsilon = 14.3 \text{ meV}$ transition.

It should be noted that in underdoped compounds the temperature dependence of $^{17}(T_1 T)^{-1}$ continuously decreases with T and at T_c only a small change in the slope can be seen[11].

5 Conclusions

We have shown that in the insulating state the main contribution to the line broadening of $4f$ transitions of Tm ions arises from the magnetic subsystem of Cu ions. The temperature dependence of the linewidth is described in terms of the linear spin-wave theory with a rather weak interaction of $4f$ levels with the magnetic moments in CuO_2 planes.

In the superconducting state due to the small coherence length of AF fluctuations, the direct contribution of AF fluctuations to the broadening of transitions is small. The temperature dependence of the linewidth is determined by the temperature dependence of the uniform static susceptibility. Note that the coupling of $4f$ levels with p -holes is weak, and due to this fact, the temperature dependence of the linewidth of $4f$ transition and the spin-lattice relaxation rate $^{89}(T_1^{-1})$ do not show any peculiarities at $T = T_c$.

In general it can be said that the inelastic scattering of neutrons provides rich possibilities for investigation of the magnetic properties of HTSC. The specific feature of the given system consists in the fact that one observes strong correlation between magnetic and phonon excitations. Therefore in the study of an excitation spectrum, it is the measurements using polarized neutrons that give the most reliable information. These measurements show, that together with magnetic fluctuations there exists a noticeable contribution of phonons.

Concerning investigation of the AF subsystem itself in the $Cu - O$ plane, the NMR method is the most efficient complemented with the method of inelastic neutron scattering on excitations in crystal fields. Such measurements confirm the existence of the antiferromagnetic Fermi-liquid suggested in[14].

Acknowledgements

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