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ON THE ROLE OF ANTIFERROMAGNETIC FLUCTUATIONS IN TEMPERATURE DEPENDENCE OF LINEWIDTH OF THE TRANSITION BETWEEN THE CRYSTAL FIELD LEVELS IN HIGH-T_c SUPERCONDUCTORS

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The problem of the role of magnetic fluctuations in the appearence of a superconducting state in high-T_c superconductors has been widely discussed. In order to understand properly the microscopic nature of the superconducting state, experimental investigation of spin dynamics in the CuO_2 planes seems to be of great importance. In particular the results of inelastic neutron scattering and nuclear magnetic resonance (NMR) experiments are of significance. In the present paper we analyze the results of inelastic neutron scattering experiments on crystal field (CF) excitations in Tm substituted $YBaCuO$ and discuss these in connection with the NMR results obtained for ^{89}Y, ^{63}Cu and ^{17}O.

Measurements of the transition linewidth between the $4f$-levels of Tm ions split by the CF of $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_x$ show a decrease in the transition width, γ, with falling temperature [1, 2]. The temperature dependence does not show any peculiarities at $T = T_c$. The authors claim that for $T^* > T_c$ the temperature dependence of the linewidth changes its behaviour [1, 2]. For a nonsuperconducting compound with x=6.1 the linewidth expands with increasing temperature. Note that the line shape is symmetrical which means that the interaction of the $4f$-levels of Tm with the excitations in CuO_2 is rather weak.

According to ref.[3] the transition linewidth between levels split by CF is determined by the dynamical susceptibility, $\chi(\epsilon)$, (ϵ is the level splitting).

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

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 $\omega \to 0$ the spin lattice relaxation rate T_1^{-1} is proportional to the dynamical susceptibility . This 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 in a wide enough temperature region [4]: $^{89}(T_1^{-1})$ shows a linear temperature dependence in nonmetallic compounds [5], $^{63}(T_1^{-1})$ does not obey the Korringa law in any temperature region. Note that for underdopped compounds, $^{63}(T_1^{-1})$ does not exhibit any peculiarities at $T = T_c$. Moreover, $^{63}(T_1^{-1})$ has a typical temperature $T^* \sim 150K$ [4], connected with antiferromagnetic fluctuations in CuO_2 planes.

Since the spin-lattice relaxation rate is determined by the imaginary part of the dynamical susceptibility, $Im\chi(q,\omega)$, at small frequencies, $\omega \to 0$, so, due to large increases of χ for large wavevectors q connected with AF fluctuations [6], the spin-lattice relaxation rates can be essentially different for different form-factors $F(q)$. According to [4, 7, 8] $^{63}(T_1^{-1})$ is determined by this susceptibility, when the AF wavevector $Q = (\pi, \pi)$, and the contribution 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 Knight shift need to be known. The main findings of the susceptibility and Knight shift measurements are as follows: i) ΔK is temperature dependent and this temperature dependence corresponds to the

temperature dependence of χ [5]; ii) the ratio $\Delta K/\chi$ does not depend on oxygen content for $6.41 < x < 7$ [5]; and iii) the temperature dependence of $\chi(T)$ changes slowly in this oxygen content region.

This paper analyzes the temperature dependence of the $4f$ transitions of Tm ions due to their CF splitting in $YBaCuO$ compounds. For the insulating state $x = 6.1$ we adopt the linear spin-wave approximation for describing the linewidth and for the superconducting compounds $YBa_2Cu_3O_{6.9}$ and $YBa_2Cu_4O_8$ we use the phenomenological approach [8].

1. $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.1}$

$Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.1}$ is an AF insulator and the main contribution to the broadening of transitions in CF arises from magnetic fluctuations. To describe $4f$ transitions we introduce the Hamiltonian similar to ref. [7]:

$$H = D\sum_{k=1}^{8} S_k\sigma \tag{2}$$

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 neighboring Cu ions. We also adopt the anisotropic Heisenberg Hamiltonian for describing the spin dynamics in CuO_2 [6]:

$$H_1 = \sum_{i,j}(JS_i^zS_j^z + J_\perp/2(S_i^+S_j^- + S_i^-S_j^+))\,. \tag{3}$$

The excitation spectrum of (3) has the gap $\Delta = J\sqrt{1-\alpha^2}$, where $\alpha = J_\perp/J$. Note that according to [6], $\Delta \sim 16 - 40meV$ in the superconducting state. In the pure AF state the gap is sufficiently small, $\Delta < 5meV$. In this latter case the temperature dependent contribution to the linewidth is determined by two processes. For $\Delta > \epsilon$ the main contribution arise from the Compton processes which describes the linewidth in the superconducting state. Note that according to [7] the spin-lattice relaxation rate can be described by the Compton processes too. For $\Delta < \epsilon$ the main contribution arise 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.

$$\gamma_{\Delta<\epsilon} = 2\pi D^2 f(\epsilon)(2n(\epsilon)+1)|u(\epsilon)-v(\epsilon)|^2 \tag{4}$$

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

Calculations of the linewidth using eq.(4) (see Fig.1) 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 $\gamma_0 = 0.17meV$ and $D = 0.03J \sim 4meV$. Moreover, the best agreement with experiment requires a small value for the spin gap, $\Delta < 5meV$. That value is in good agreement with the experimental data reported in [6].

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.

2. $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.9}$

To analyze the temperature dependence of the linewidth in superconducting compounds we use the phenomenological approach [8], and the approximation for $\chi(q,\omega)$ as discussed in [4].

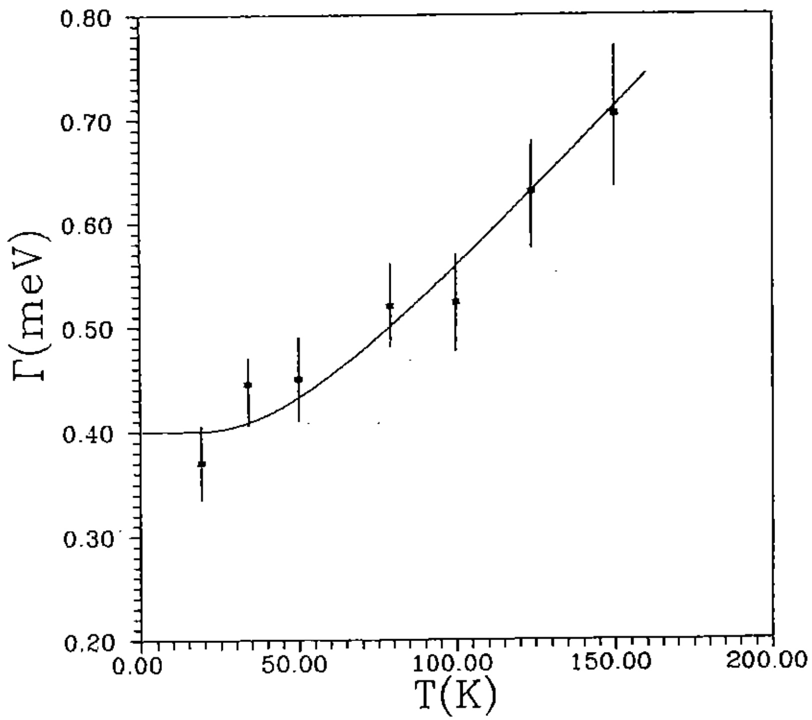

Figure 1: Temperature dependence of the linewidth in insulating state $x = 6.1$.

According to the phenomenological theory of an antiferromagnetic Fermi liquid [8], the dynamical susceptibility for small frequencies $\omega \rightarrow 0$ has the form:

$$\chi(q,\omega) = \frac{\pi\omega\chi_0(T)}{\Gamma(T)}\left[1+\frac{\beta(\xi/a)^4}{1-\xi^2(Q-q)^2}\right], \tag{5}$$

where $\chi_0(T)$ is the uniform susceptibility, Γ is the typical energy of the quasiparticle spectrum of spin fluctuation, β is the parameter characterizing the weight of the AF component of susceptibility, and ξ is the correlation length of AF fluctuation. The temperature dependence of the correlation length has the form:

$$(\frac{\xi(T)}{a})^2 = (\frac{\xi_0}{a})^2(\frac{|T_x|}{T+T_x}). \qquad (6)$$

Note that the investigation of AF correlation in superconducting materials [6] does not provide for such type of dependence. However for the analysis of temperature dependence of the $4f$ transition linewidth, the contribution of $\chi(Q)$ is suppressed due to the fact that $F(Q) = 0$ and temperature dependence of the correlation length is not essential.

Also note that according to ref. [4] the squared Lorentzian proposed in [8] for **Q** dependence of the susceptibility (5) is definitely too wide to explain the temperature dependence of $^{17}(T_1T)^{-1}$. According to the ref. [9] the **Q** dependence of susceptibility is described by a Gaussian and falls off much faster than a Lorentzian **Q** dependence. According to ref.[4] the dynamical susceptibility can be approximated by the following expression (which is supported by the neutron scattering experiments [9]):

$$\chi(q) = \frac{\pi}{4}\sum_{i=1}^{4}\frac{\chi_0(T)+\chi(Q^i)\phi(Q^i-q)}{\Gamma(T)+(\Gamma(T)-\Gamma_{SF})\phi(Q^i-q)}, \qquad (7)$$

where $\phi(q) = exp(-ln2\xi^2q^2)$, $Q^i = (\pi\pm\delta,\pi),(\pi,\pi\pm\delta)$, $\chi(Q^i)$ is the susceptibility for the AF wavevector, Γ_{SF} is the typical energy of spin fluctuation for Q^i. Note that (7) takes into account the appearance of incommensurate AF fluctuation under the dopping of CuO_2 planes by holes. Note that the **Q** width of the AF Gaussian peak of susceptibility $\chi(Q,\omega)$, according to ref. [9], 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. [6]

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 (7) 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 [4]. Indeed, the contribution of the second term in eq. (7) can be easily estimated as

$$1/(16\pi(ln2\xi^2)^3)\cdot\chi(Q_{AF})/\Gamma(T). \qquad (8)$$

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

$$\gamma \sim \frac{\epsilon D^2\chi_0(T)}{\Gamma(T)}cth(\epsilon/2T). \qquad (9)$$

To analyze the temperature dependence of linewidth we use eq. (9) and the temperature dependence of $\chi_0(T)$ and $\Gamma(T)$ as presented in ref. [8]. The best agreement between theoretical and experimental results (Fig. 2) corresponds to the following set of parameters: $D \sim 5meV$, $\gamma_0 \sim 0.15meV$.

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 [1, 2]. However recent experimental results obtained with $Tm_{0.1}Y_{0.9}Ba_2(Cu_{1-x}Zn_x)_3O_{6.9}$ and $Tm:YBa_2Cu_4O_8$ [10, 11] do not show any peculiarities for $T > T_c$ within experimental error. It should be noted that in underdopped compounds the temperature dependence of $^{17}(T_1T)^{-1}$ continuously decreases with T and at T_c only a small change in the slope can be seen [4]. Fig. 3 shows the results of the linewidth calculation of $4f$ transition in Tm substituted $YBa_2Cu_4O_8$ in comparison with the experimental data [11]. The temperature dependence, $\chi(T)$, is presented in ref. [12]. The best agreement of the theoretical and experimental results

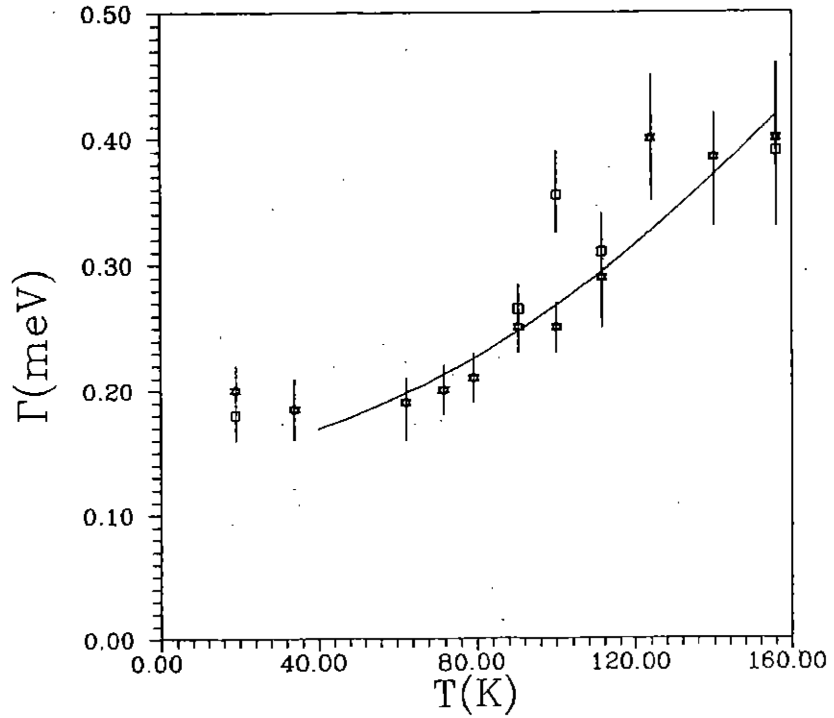

Figure 2: Temperature dependence of the linewidth in superconducting state $x = 6.9$.

corresponds to the following set of the parameters: $D \sim 6meV$, $\gamma_0 = 0.27meV$ for the $\epsilon = 11.8meV$ transition and $\gamma_0 = 0.31meV$ for the $\epsilon = 14.3meV$ transition.

Therefore, 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 superconducting compounds due to the small coherence length of AF fluctuation, the direct contribution of AF fluctuation to the broadening of

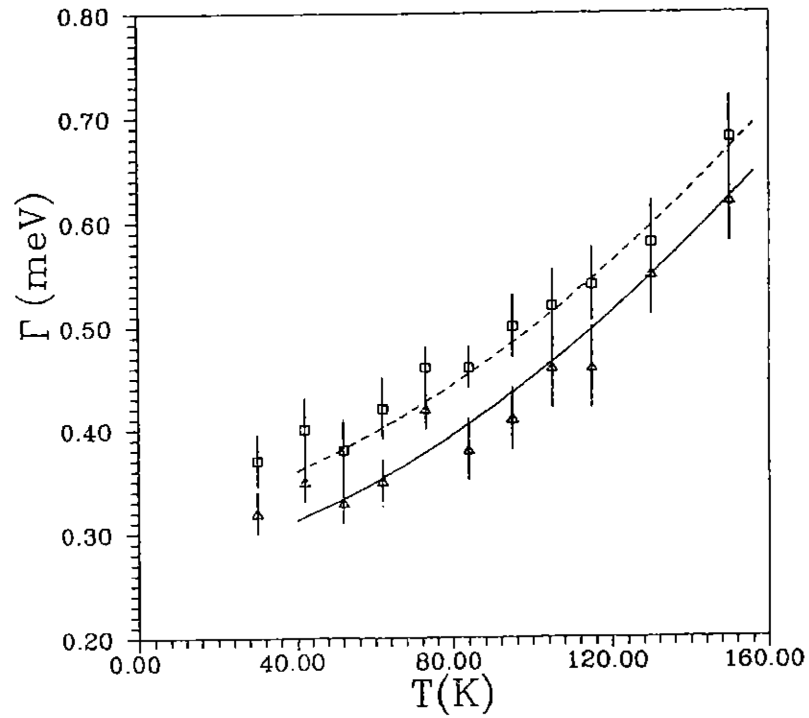

Figure 3: Temperature dependence of the linewidth in $YBa_2Cu_4O_8$. (- - corresponds to $\epsilon = 11.8$ meV, - - corresponds to $\epsilon = 14.3$ meV.

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 $T = T_c$.

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