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Role of antiferromagnetic fluctuations in the temperature dependence of the linewidth of the transitions between the crystal-field levels in high-T_c superconductors

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The measurements of linewidth of the transitions between 4f levels of Tm split by the crystal field in Y-Ba-Cu-O compounds are analyzed using the spin-wave approximation and the phenomenological approach of Millis, Monien, and Pines. It was shown that in the insulating state the main contribution to the linewidth arises from one-magnon processes and a gap in the spectrum of magnetic excitation is rather small ($\Delta < 100$ K). In superconducting compounds the main contribution to the linewidth arises from zone-center magnetic fluctuations and the temperature dependence of the linewidth is determined by static spin susceptibility.

Magnetic fluctuations as a mechanism of high-T_c are now 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 Y-Ba-Cu-O and discuss these in connection with the NMR results on ^{86}Y, ^{63}Cu, and ^{17}O.

Measurements of the linewidth of the transition between 4f levels of Tm ions split by the CF of $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_x$ show a decrease of the transition width γ with decrease of 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 behavior.[1,2] For the nonsuperconducting compound with $x = 6.1$, the linewidth grows with increasing temperature. Note that the line shape is symmetric. It means that the interaction of the 4f levels of Tm with excitations in CuO$_2$ is rather weak.

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

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

where F_q is the geometrical form factor corresponding to the ionic position in a unit cell. According to Eq. (1), the linewidth is proportional to the imaginary part of the self-energy of f electrons. The imaginary part of self-energy is not equal to zero due to spin-spin interaction and is proportional to the $\langle S^-(t)S^+(0)\rangle$ correlator.[4]

In the case of Y-Ba-Cu-O, Eq. (1) describes the linewidth of the CF transition caused by both interaction with excitation in a subsystem of local spins on Cu ions in CuO$_2$ planes, and the interaction with conducting p holes. We suppose here that the main contribution to the linewidth comes from spin-spin interaction of f electrons and local spins on the Cu sublattice. This is supported by the fact that broadening of the transitions in the insulating antiferromagnetic (AF) state $YBa_2Cu_3O_{6.1}$ is two times larger than in the superconducting state. It is connected with a decrease in the magnetic correlation length in the superconducting phase and weak interaction of f electrons with p holes.

Note that the spin-lattice-relaxation rate T_1^{-1} is proportional to the integral of the imaginary part of dynamic susceptibility for $\omega \to 0$, with filtering form factor $F(q)$. That means that for small splitting ϵ of 4f levels, the linewidth of the CF transition and spin-lattice-relaxation rate should show similar temperature dependencies.

The spin-lattice-relaxation rates for ^{89}Y and ^{17}O obey the Korringa law in a wide enough temperature region.[5] $^{89}(T_1^{-1})$ shows the linear temperature dependence in nonmetallic compounds.[6] $^{63}(T_1^{-1})$ does not obey the Korringa law in any temperature region. Note that for underdoped compounds $^{63}(T_1^{-1})$ does not show peculiarities for $T = T_c$. Moreover, $^{63}(T_1^{-1})$ has a typical temperature $T^* \sim 150$ K,[5] connected with antiferromagnetic fluctuations in CuO$_2$ planes.

Since the spin-lattice-relaxation rate is determined by the imaginary part of dynamical susceptibility $\mathrm{Im}\chi(q,\omega)$ at small frequencies $\omega \to 0$, so due to a large increase of χ for large wave vectors q connected with AF fluctuations,[7] the spin-lattice-relaxation rates can be essentially different for different form factors $F(q)$. According to Refs. 5, 8, and 9 $^{63}(T_1^{-1})$ is determined by the susceptibility at the AF wave vector $Q = (\pi,\pi)$, but the contribution of $\chi(Q)$ to relaxation rates of ^{17}O and ^{89}Y is small, due to the fact that form factor $F(Q) = 0$. In the last case, the spin-lattice-relaxation times are determined by $\chi(q = 0)$.

For the purpose of understanding the temperature dependence of relaxation rates and 4f-transition widths, it is important to know the temperature dependence of the static susceptibility and the Knight shift. The main findings of the susceptibility and Knight shift measurements are as follows: (i) ΔK is temperature dependent

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and this temperature dependence corresponds to the temperature dependence of χ,[6] (ii) the ratio $\Delta K/\chi$ does not depend on oxygen content for $6.41 < x < 7$.[6]

We analyze in this paper the temperature dependence of $4f$ transitions of Tm ions due to their CF splitting in Y-Ba-Cu-O compounds. In the insulating state $x = 6.1$, we adopt the linear spin-wave approximation for describing the linewidth. For superconducting compounds we use the phenomenological approach.[9]

Linewidth in insulating AF phase

$Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.1}$ is an AF insulator and the main contribution to broadening of transition in CF arises from magnetic fluctuations. To describe $4f$ transitions, we introduce the Hamiltonian similar to Ref. 8,

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

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 eight nearest-neighboring Cu ions. We also adopt the anisotropic Heisenberg Hamiltonian for describing the spin dynamics in CuO_2:[7]

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

The excitation spectrum of (3) has the gap $\Delta = J\sqrt{1-\alpha^2}$, where $\alpha = J_\perp / J$. Note that according to Ref. 7, $\Delta \sim 16-40$ meV in the superconducting compounds. In the pure AF state, the gap is sufficiently small $\Delta < 5$ meV. In that case, the temperature-dependent contribution to linewidth is determined by two processes. For $\Delta < \epsilon$, the main contribution arises from Compton processes. These processes describe the linewidth in the superconducting compounds. Note that according to Ref. 8 the spin-lattice-relaxation rate is determined by Compton processes, too. For $\Delta < \epsilon$, the main contribution arises from one-magnon emission absorption processes. On the other hand, the spin-lattice-relaxation time is also determined by 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

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

Calculations of the linewidth using Eq. (4) (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 $\gamma_0 = 0.17$ meV and $D = 0.03J \sim 4$ meV. Moreover, the best agreement with experiments requires a small value of spin gap $\Delta = 3$ meV < 5 meV. That value is in good agreement with experimental data reported in Ref. 7.

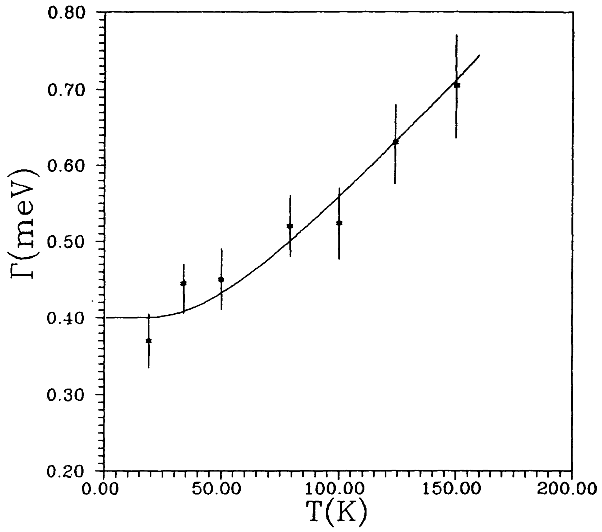

FIG. 1. Temperature dependence of the linewidth in insulating state $x = 6.1$

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

Linewidth in superconducting phase

To analyze the temperature dependence of the linewidth in superconducting compounds, we use the phenomenological approach,[9] and approximation for $\chi(q,\omega)$ as discussed in Ref. 5.

According to the phenomenological theory of antiferromagnetic Fermi liquid,[9] 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 the susceptibility, and ξ is the correlation length of AF fluctuation. The temperature dependence of the correlation length has the following form:

$$\left[\frac{\xi(T)}{a}\right]^2 = \left[\frac{\xi_0}{a}\right]^2 \left[\frac{|T_x|}{T+T_x}\right] . \tag{6}$$

Note that the investigation of AF correlation in superconducting materials[7] does not provide such a 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 the temperature dependence of the correlation length is not essential.

Note that according to Ref. 5, the squared Lorentzian proposed in Ref. 9 for **Q** dependence of the susceptibility (5) is definitely too wide to explain the temperature dependence of $^{17}(T_1T)^{-1}$. According to Ref. 10, **Q** dependence of susceptibility is described by a Gaussian and falls off much faster than a Lorentzian **Q** dependence. According to Ref. 5, the dynamical susceptibility can be approximated by the following expression (which is supported by neutron-scattering experiments):[10]

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

where $\phi(q)=\exp(-\ln 2\xi^2q^2)$, $Q^i=(\pi\pm\delta,\pi),(\pi,\pi\pm\delta)$, $\chi(Q^i)$ is the susceptibility for the AF wave vector, and 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 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. 10 is ω dependent, so that straightforward determination of the correlation length is impossible. A rough estimate shows that $\xi\sim 2-3a$, which is in agreement with Ref. 7.

Equation (1) for the linewidth of the $4f$ transition contains form factor $F(Q)$, which is equal to 0 for $q=Q$. It means that the contribution of the second term of Eq. (7) is small. According to estimates from absolute values of spin-lattice-relaxation rates on $^{63}(T_1^{-1})$ and $^{17}(T_1^{-1})$, it is less than 0.01.[5] Indeed, the contribution of the second term in Eq. (7) can be easily estimated as

$$1/[16\pi(\ln 2\xi^2)^3]\chi(Q_{\mathrm{AF}})/\Gamma(T)\ . \tag{8}$$

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 wave vector is strongly suppressed,

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

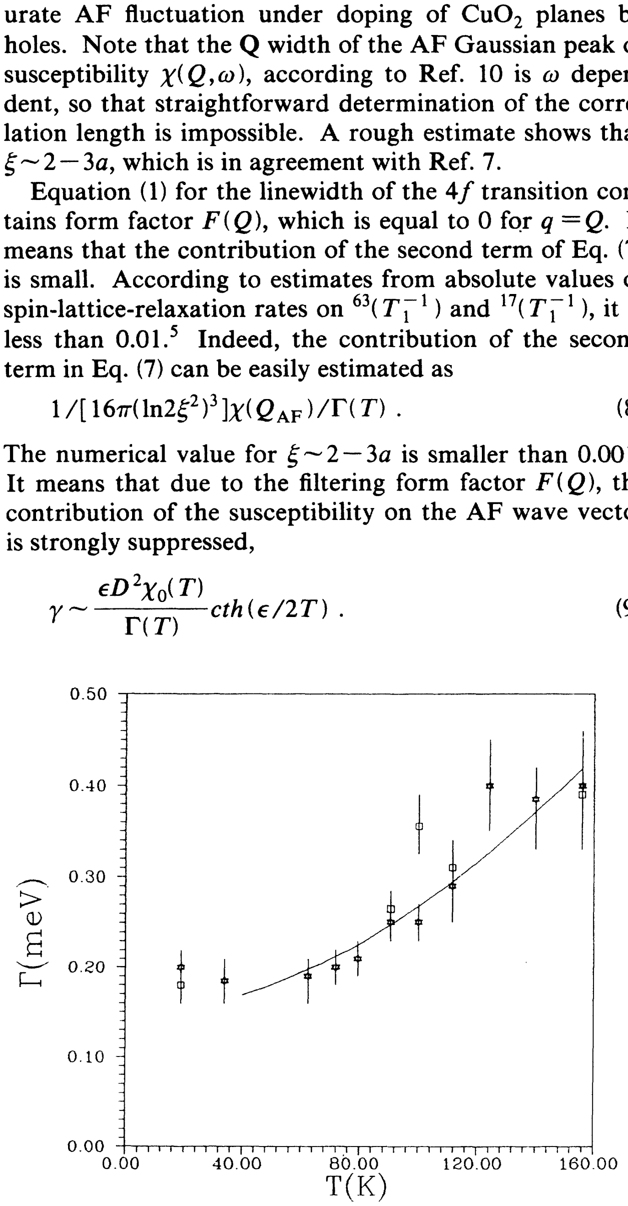

FIG. 2. Temperature dependence of the linewidth in superconducting state $x=6.9$.

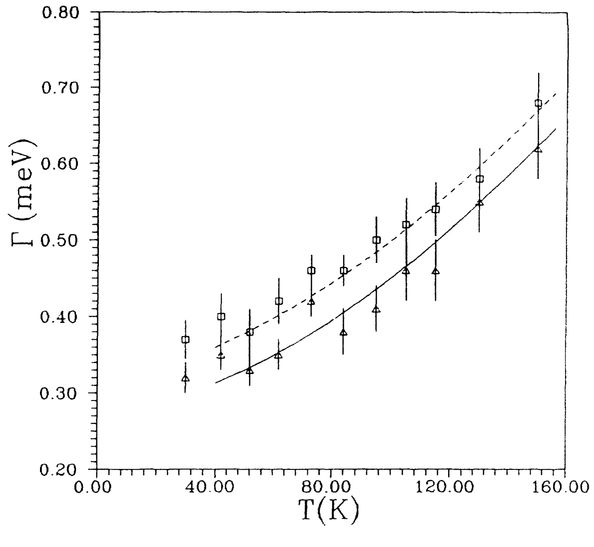

FIG. 3. Temperature dependence of the linewidth in $YBa_2Cu_4O_8$, (solid line) corresponds to $\epsilon=11.8$ meV, (dashed line) corresponds to $\epsilon=14.3$ meV.

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

It is worth mentioning that the model proposed for describing linewidth in high-T_c superconductors does not lead to the appearance of any peculiarities at $T^*>T_c$, which was discussed in Refs. 1,2. However, recent experimental results on $Tm_{0.1}Y_{0.9}Ba_2(Cu_{1-x}Zn_x)_3O_{6.9}$ and $(Tm/Y)Ba_2Cu_4O_8$ (Refs. 11,12) do not show peculiarities for $T>T_c$ within experimental error. It should be noted that in underdoped 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.[5] Figure 3 shows the results of the calculation of the linewidth of the $4f$ transition in Tm-substituted $YBa_2Cu_4O_8$ and comparison with the experimental data.[12] The temperature dependence of $\chi(T)$ is presented in Ref. 13. The best agreement of the theoretical and experimental results corresponds to the following set of parameter: $D\sim 6$ meV and $\gamma_0=0.27$ meV for the $\epsilon=11.8$ meV transition, and $\gamma_0=0.31$, meV for the $\epsilon=14.3$ meV 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 linear spin-wave theory with rather weak interaction of $4f$ levels with magnetic moments in CuO_2 planes.

In superconducting compounds, the direct contribution of AF fluctuations on the broadening of transitions is small. The temperature dependence of the linewidth is determined by 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 the $4f$ transition and spin-lattice-relaxation rate ${}^{89}(T_1^{-1})$ do not show any peculiarities at $T=T_c$.

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