Effect of spin fluctuations on the linewidth of 4f-electron transitions in Tm–YBCO compounds

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The *s–f* interaction of the rare-earth ion Tm^{3+} with copper spins in Tm–YBCO compounds is analyzed. An expression is derived for the linewidth of transitions in the crystal electric field for Tm^{3+}. Dynamic spin fluctuations in CuO_2 dominate the line broadening.

In some recent experiments[1,2] by inelastic neutron scattering, a study was made of the temperature dependence of the linewidth of transitions between the ground level $\Gamma_3^{(1)}$ and the first excited levels $\Gamma_4^{(1)}$ and $\Gamma_2^{(1)}$ of the crystal electric field for Tm^{3+} ions in the high-T_c superconductor $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.9}$ (Tm–$YBCO_{6.9}$) at energies of 11.8 and 14.2 meV, respectively. It was found that the linewidth decreases sharply at temperatures $T_S \simeq T_c + 20$ K ($T_c = 92$ K) for both transitions. For the transition between the ground level and the first excited level, with an energy $\delta = 13.8$ meV for the nonsuperconducting compound Tm–$YBCO_{6.1}$, Osborn and Goremychkin[2] approximated this dependence by the expression $\Gamma \propto \mathrm{Im}\,\chi(\delta)\cdot\coth(\delta/2kT)$, under the assumption that the imaginary part of the magnetic susceptibility χ was independent of the temperature. It can be seen from the experimental data that the values of the linewidth Γ for Tm–$YBCO_{6.9}$ are smaller at all temperatures than the corresponding width for Tm–$YBCO_{6.1}$. The implication is that the Fermi-liquid contribution to the relaxation of the magnetic excitations of the Tm^{3+} f ions in the crystal electric field is small.

The long-range antiferromagnetic order of the spins at the Cu2 sites would lead us to expect the spin–spin interaction of *f* electrons and the antiferromagnetic spin waves to be the primary cause of the broadening in Tm–$YBCO_{6.1}$. With increasing hole density in the CuO_2 layer, and with the delocalization of holes, there is a significant decrease in the magnetic correlation length, and the long-range order disappears. However, strong antiferromagnetic correlations of the spins are retained in the superconducting and metallic phases.[4–6] One might thus expect that spin fluctuations in the CuO_2 planes would provide the primary mechanism for the relaxation of the excitations of *f* electrons, leading to a broadening of the line of the crystal electric field in Tm–$YBCO_{6.9}$. In this letter we calculate the temperature dependence of the width of the excitation lines in the crystal electric field which stem from dynamic spin fluctuations in $YBCO_{6.9}$.

1. The model. In light of the NMR experiments,[4] we consider a model of an antiferromagnetic Fermi liquid at each Cu2 site of which the spin is $S = 1/2$.

We write the Hamiltonian of the rare-earth ions and the spins $S = 1/2$ at the copper sites as follows:

$$H = H_{CEF} + H_{t\text{-}J} + H_{S\text{-}F}\,, \tag{1}$$

$$H_{CEF} = \sum_{n,i} \omega_n K_{nn,i}\,, \tag{2}$$

where $K_{mn,i} = (|m\rangle\langle n|)_i$ are the transition operators for the n,m levels of the crystal electric field,

$$H_{t\text{-}J} = -t \sum_{i,j} \hat{c}^{+}_{i\sigma}\hat{c}_{j\sigma} + J \sum_{i,j} \mathbf{S}_i \mathbf{S}_j \tag{3}$$

is the Hamiltonian of the t–J model describing holes $\hat{c}^{+}_{i\sigma} = \hat{c}^{+}_{i\sigma}(1-n_{i,-\sigma})$ in the CuO_2 plane and the interaction of the spins $\mathbf{S}_i$ at the copper sites, and

$$H_{S\text{-}F} = -\sum_{i,j} I^{\alpha}_{S\text{-}F} J^{\alpha}_i S^{\alpha}_j\,, \tag{4}$$

where the operator $\mathbf{J}_i$ represents the total angular momentum of the Tm^{3+} ion at site i, the operator $\mathbf{S}_j$ represents the spin at the copper sites j nearest this site, and $I^{\alpha}_{S\text{-}F}$ is the indirect-exchange coupling constant. The Hamiltonian of the S–F interactions in (4) has a form close to that of the Hamiltonian representing the interaction of conduction electrons with $4f$ electrons.[3,7]

Using the technique of differentiating with respect to two times in the equations-of-motion method for the two-time Green's functions $\langle\langle J^{-}_i(t)\,|\,J^{+}_{i'}(t')\rangle\rangle$, we can easily find the mass operator $\Sigma_i(\omega)$ of the Dyson equation. The level width $\Gamma_i(\omega)$ is determined by the imaginary part of the mass operator:

$$\Gamma_i(\delta) = -\coth\left(\frac{\delta}{2kT}\right)\mathrm{Im}\left\{\sum_i (\delta + i\varepsilon)\right\}, \tag{5}$$

where

$$\sum_i (\omega) = \frac{1}{4} \sum_{j,j'} \tilde{I}_{ij} \langle\langle S^{-}_j \,|\, S^{+}_{j'}\rangle\rangle_\omega \, \tilde{I}_{ij'}\,, \tag{6}$$

the summation is over the jj' copper sites in the neighboring CuO_2 planes, and $\tilde{I}_{ij} = \tilde{I}(i-j) \propto I^{xx}_{S\text{-}F} \approx I^{yy}_{S\text{-}F}$.

Going over to the q representation, we find

$$\Gamma(\delta) \propto \coth\left(\frac{\delta}{2kT}\right) \sum_{\mathbf{q}} [F(\mathbf{q})]^2 \cdot \mathrm{Im}\{\chi^{-+}(\mathbf{q},\delta)\}, \tag{7}$$

where

$$F(\mathbf{q}) = 8\cos\left(\frac{aq_x}{2}\right)\cos\left(\frac{bq_y}{2}\right)\cos\left(\frac{cq_z}{6}\right) \tag{8}$$

is a form factor[4] which incorporates the local symmetry of the Tm^{3+} ion in the $TmBa_2Cu_3O_7$ unit cell with the lattice constants a, b, c; and

$$\mathrm{Im}\{\chi^{-+}(\mathbf{q},\omega)\} \equiv \chi''_{-+}(\mathbf{q},\omega) = -\langle\langle S^{-}\,|\,S^{+}\rangle\rangle_{\mathbf{q},\omega} \tag{9}$$

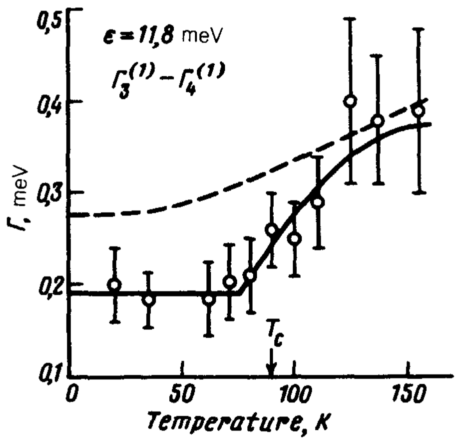

FIG. 1. Temperature dependence of the width of a transition between the ground level $\Gamma_3^{(1)}$ and the first excited level $\Gamma_4^{(1)}$ for an energy $\varepsilon=11.8$ meV of the Tm^{3+} ion in Tm–$YBCO_{6.9}$. Circles—Experimental data; solid line—theoretical; dashed line—$\Gamma_0[1+\coth(\varepsilon/2kT)]$.

is the imaginary part of the Fourier component of the complex generalized susceptibility associated with the corresponding retarded Green's function.

Rossat-Mignod *et al.*[5] have recently measured the temperature dependence of the imaginary part of the magnetic susceptibility of $YBCO_{6.92}$ near the antiferromagnetic wave vector $\mathbf{Q}_{AF}=(1/2;\ 1/2;\ 1,\ 6)$ for an energy $\hbar\omega=10$ meV. This dependence can be approximated accurately at $T>75$ K by

$$\chi''(\omega,T)\simeq a\cdot(T-75)^b\cdot\exp[c(T-75)], \tag{10}$$

where the parameters have the values

$a=1.22\,K^{-b}$, $b=1.148$, and $c=-0.017\,K^{-1}$.

Since the average spin susceptibility over the vectors $\mathbf{q}$ near Q_{AF} is measured experimentally, we can assume that the temperature dependence of the sums over $\mathbf{q}$ in (7) corresponds to (10), i.e.,

$$\sum_{\mathbf{q}}[F(\mathbf{q})]^2\cdot\mathrm{Im}\{\chi^{-+}(\mathbf{q},\delta)\}\propto\chi''(\omega,T). \tag{11}$$

Assuming that the linewidth has a constant component Γ_0 which is not due to dynamic spin fluctuations, we find the following expression for the linewidth:

$$\Gamma(T)=\Gamma_0+A\cdot\coth\left(\frac{\delta}{2kT}\right)\chi''(\omega,T), \tag{12}$$

where a fit of (12) to the experimental data[2] yielded the parameter values $\Gamma_0=0.187$ meV and $A=0.172\times10^{-2}$ [in corresponding units].

The circles in Fig. 1 show experimental results on the linewidth for the $\Gamma_3^{(1)}\rightarrow\Gamma_4^{(1)}$ transition with an energy $\delta=11.8$ meV in Tm–$YBCO_{6.9}$. The dashed curve shows a plot of $\coth(\delta/2kT)$, while the solid curve shows results calculated from (12).

A sharp decrease in the linewidth is also observed in $Tm_{0.1}Y_{0.9}Ba_2(Cu,Zn)_3O_{6.9}$ at $T_S\simeq T_c+20$ K, where we have $T_c=50$ K at a Zn concentration of 5%.

This model thus explains the temperature dependence of the transition linewidth for $4f$ electrons in the crystal electric field. The sharp decrease in the linewidth at $T_S \simeq T_c + 20$ K $> T_c$ indicates that dynamic spin fluctuations contribute substantially to the line broadening. According to NMR experiments,[6] these fluctuations reach a maximum at the temperature T_S. In this regard, these materials differ from conventional metals, e.g.,[3] $Tb_xLa_{1-x}Al_2$. In this case the decrease in the linewidth occurs at $T = T_c$ because of the formation of a superconducting gap $2\Delta < \delta_{CF}$.

We intend to carry out in a separate paper a more systematic calculation of linewidth (7), using analytic expressions for $\chi''(\mathbf{q},\omega)$ according to the t–J model.

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