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INFLUENCE OF THE SUPERCONDUCTING ENERGY GAP ON THE LINE WIDTH OF THE CRYSTAL FIELD TRANSITION IN THE HIGH T_c SUPERCONDUCTOR $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.9}$

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Relaxation of localized 4f magnetic moment in metallic rare earth (RE) systems caused by interaction with conduction electrons (s-f interaction) manifests itself in the broadening of peaks in spectra of inelastic neutron scattering (INS) on transitions between the levels of a splitted by crystalline electric field (CEF) ground multiplet of an RE ion[1]. In a superconducting state this mechanism of decay of CEF excitations may lead to the breaking off a Cooper pair (spin flip of one of the electrons forming the Cooper pair $(\vec{k}\uparrow, -\vec{k}\downarrow)$) if the energy of a CEF transition (ε) exceeds the superconducting energy gap (SEG) ($2\Delta(T)$). An appearance of the SEG at $2\Delta(T) < \varepsilon$ does not affect to the first approximation the temperature dependence of the line width of the CEF excitation. The situation is different for $2\Delta(T) > \varepsilon$, when the CEF excitation energy ε is insufficient for a Cooper pair to break. As a result the relaxation channel caused by s-f interaction will be switched off leading to a rapid exponential decrease of the line width at $T < T_c$[2].

In this Letter we report on the results of the investigation of the temperature dependence of the line width of CEF transitions by means of the INS technique in a high T_c superconductor $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.9}$ (Tm:YBCO). The J ground state multiplet of the Tm^{+3} ion in an orthorhombic CEF splits into 13 singlets Γ_i: $4x\Gamma_1$, $3x\Gamma_2$, $3x\Gamma_3$, $3x\Gamma_4$. The lowest levels are the Γ_3 ground state followed by a Γ_4 at 11.8 meV and a Γ_2 at 14.2 meV. Between these lowest levels two ground state transitions are allowed. This circumstance makes the Tm:YBCO system very suitable for the investigation of the temperature dependence of the line width of CEF transitions, because the INS spectra contain two strong, weakly overlapping inelastic peaks at the energy transfer 11.8 meV and 14.2 meV. A 10% substitution of Y for Tm excludes influence of the 4f-4f interaction on the line width of CEF transitions. According to X-ray and neutron diffraction measurements the Tm:YBCO sample was single-phase. The onset superconductivity temperature was determined to be $T_c = 92K$ with $\Delta T_c = 2K$.

Inelastic neutron scattering experiments were performed using a direct geometry chopper spectrometer HET[3] at the pulsed spallation source ISIS, Rutherford Appleton Laboratory, U.K. The incoming energy of E_0 = 35 meV was used, at a resolution on elastic line of 0.3 meV (HWHM) as derived from vanadium spectrum. The mean scattering angle is $\varphi = 5^\circ$ which corresponds to a momentum transfer ranging from 0.6 $Å^{-1}$ to 1.12 $Å^{-1}$ within an energy transfer interval from 8 meV to 16 meV. A 100 g of Tm:YBCO sample in a thin-wall aluminium can, filled with helium gas to provide better thermal contact was mounted on a cold finger of a closed cycle refrigerator giving a temperature from 15 K to 300 K.

Fig.1 gives as an example three fragments of INS spectra of Tm:YBCO at 125 K (above T_c), 90 K (near T_c) and 35 K (below T_c). In experimental data processing the following was taken into consideration:

a) since the peaks at ε_1 = = 11.8 meV and ε_2 = 14.2 meV are due to ground state transitions, the ratio of their intensities is equal to the ratio of square matrix elements of the $J_\perp$ operator (I_2/I_1 = 0.83) and independent of temperature;

b) at high temperatures (T > 100 K), weak contributions to INS spectra appear due to the scattering on CEF transitions from excited levels. These contributions were taken into account in accord with the full CEF level scheme;

c) the modelled scattering law as a set of Lorentzians was convoluted with the resolution function of the spectrometer. The background was fitted by a straight line beyond the interval containing the magnetic signal.

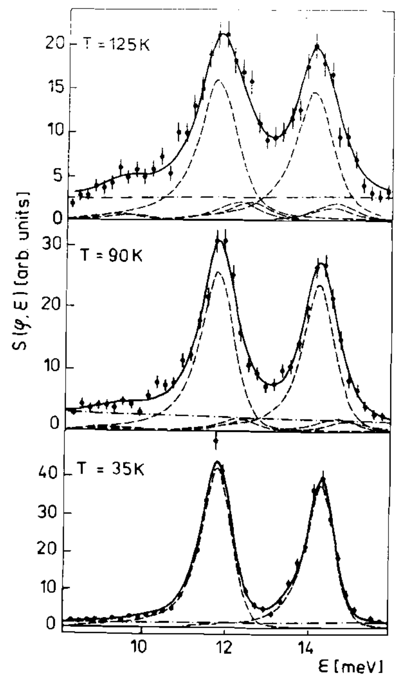


Fig.1. Fragments of INS spectra (points) for an interval of energy transfer from 8 meV to 16 meV. For details see the text.

In result of our fitting procedure we obtained three independent parameters: widths of two CEF ground state transitions and an intensity of the peak at ε_1 = 11.8 meV. In fig.1 the solid line is the least

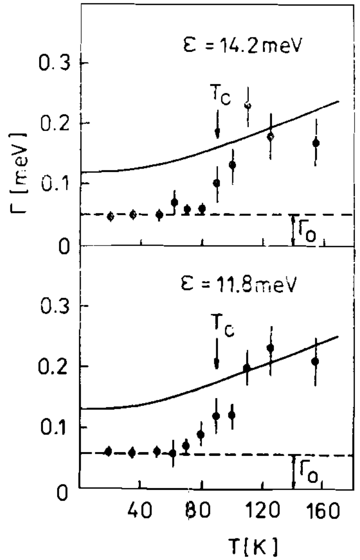
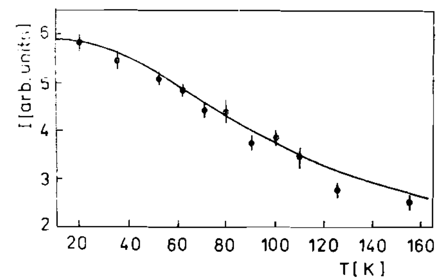


Fig.2. Temperature dependence of the HWHM of the CEF transitions at energy transfer 11.8 meV and 14.2 meV.

Fig.3. Temperature dependence of the intensity of transition at 11.8 meV(points), solid line is the calculation in the CEF model.



square fit to the experimental points, the dashed lines are the separate spectral components, the point-dashed line is the background. The results of the determination of the line widths of the CEF excitations are given in fig.2. The arrows point to the superconducting transition temperature. As shown in this figure at T < 70 K the line width is T-independent with a residual width of Γ_0 = 0.05 meV. With increasing temperature near T_c a considerable broadening of CEF transitions takes place. Such a behaviour of the width temperature dependence is due to an appearance of a channel of relaxation - the creation of electron-hole excitations at T > T_c caused by the closing of the SEG. The residual width Γ_0 may result from statical distortions due to the deficit of oxygen (δ = 0.1). In fig.3 there is shown the temperature dependence of the intensity of the CEF transition at ε_1 = 11.8 meV.Solid line is the calculation in the CEF model of the population of the ground level $\rho(T) = Z(T)^{-1}$,(Z(T) is the partition function) normalized to the experimental point at T = 20 K. Good agreement between the theoretical curve and experimental points in this figure justifies the method of data analysis.

In a normal state (T > T_c) the temperature dependence of the line width for the case of two singlets is given by/1/:

$$\Gamma = 4\pi M^2 (N(0)J_{ex})^2 (g_J - 1)^2 \delta \coth\frac{\delta}{2T}, \qquad (1)$$

where $M=\langle m|J|n\rangle$, N(0) is the density of states of conduction electrons at the Fermi level, J_{ex} is the exchange integral, g_J is the Lande factor, δ is the energy between two singlets. By fitting to expression (1) of experimental values at $T > T_c$ (solid line in fig.2) we obtained estimates for the coupling constant $\rho' = N(0)J_{ex} = 0.025\pm0.005$, which coincide with the value of ρ' for an isostructural compound $ErBa_2Cu_3O_7$ [4]. It should be noted that the value of ρ' for high T_c compounds is not negligible and its order of magnitude corresponds to ρ' for a metallic superconductor $Tb:LaAl_2$ ($T_c = 3.2$ K; $\rho' = 0.034$) [5]. In fig.2, at $T < T_c$ the experimental points are located below the calculated curve. Apparently, in a superconducting state $N(0) \approx 0$ (or at least sharply decrease in comparison with that in a normal state), if we accept that Γ_0 is not caused by the s-f interaction (to which fact the absence of a temperature dependence of the line width at $T < 70$ K indicates).

In conclusion, it should be noted, that we are making plans for more thorough measurements of the temperature dependence of the line width of the CEF excitations near T_c which could probably allow the determination of the value of the SEG (for that it is necessary to know precisely the line width behaviour below T_c). Within the our available accuracy of the experiment a lower limit of $2\Delta(0) > 14.2$ meV can be only indicated.

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Горемычкин Е.А., Осборн Р., Тейлор А.Д. E14-89-721
Влияние энергетической щели в высокотемпературном сверхпроводнике $Tm_{0,1}Y_{0,9}Ba_2Cu_3O_{6,9}$ на ширины переходов между уровнями кристаллического поля

Методом неупругого рассеяния нейтронов исследована температурная зависимость ширин переходов между уровнями кристаллического поля в высокотемпературном сверхпроводнике $Tm_{0,1}Y_{0,9}Ba_2Cu_3O_{6,9}$. При $T < T_c$ наблюдено уменьшение ширин переходов, обусловленное открытием энергетической щели при переходе в сверхпроводящее состояние. Оценена константа, характеризующая силу s-f взаимодействия $\rho' = 0{,}025 \pm \pm 0.005$.

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Goremychkin E.A., Osborn R., Taylor A.D. E14-89-721
Influence of the Superconducting Energy Gap on the Line Width of the Crystal Field Transition in the High T_c Superconductor $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.9}$

By inelastic neutron scattering the temperature dependence of the line width of crystal field transitions were measured in the high T_c superconductor $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.9}$. At $T < T_c$ the decreasing of the line width due to the opening of the superconducting energy gap was observed. The coupling constant ρ' which characterizes the strength of an s-f interaction was estimated to be $\rho' = 0.025\pm0.005$.

The investigation has beem performed at the Laboratory of Neutron Physics, JINR and in Rutnerford Laboratory, U.K.

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