Neutron scattering investigations of the antiferromagnetic phase of YBCO nano-particles

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Abstract

Undoped bulk $YBa_2Cu_3O_{6+x}$ (YBCO) is antiferromagnetically ordered with a Néel temperature of approximately 410 K. Doped bulk YBCO becomes a high T_C superconductor at doping levels $x \geq 0.35$.

In our studies we investigate nano-sized particles of YBCO and thereby use the particle size as a new control parameter to gain additional insight into the physics of YBCO. We expect the particle size to be relevant when it becomes comparable to the superconducting coherence length. So far we have investigated the antiferromagnetic phase diagram In YBCO nano-particles with mean diameter of 40 nm, and present a mapping of the antiferromagnetically ordered phase as a function of oxygen doping level.

The neutron powder diffraction investigations have been conducted using a triple axis instrument to focus on a narrow q-range and to suppress background, since the magnetic signal is very weak.

Overview

The coherence length in bulk superconducting YBCO is 3 nm in the a,b-plane and 0.4 nm along the c axis. It is believed that the mechanism behind superconductivity in YBCO is closely related to the magnetism. One of many indications is that the superconducting phase emerges at dopings above x=0.35 - the same doping, as where the antiferromagnetic order disappears in bulk YBCO.

Assuming that the superconducting and the antiferromagnetic phase are interrelated, it is interesting to investigate what happens to the antiferromagnetic order at particle sizes comparable or smaller than the coherence length for the Cooper-pairs in YBCO.

We are therefore performing a mapping of the antiferromagnetic phase, including order parameter, Néel temperature, magnetization versus temperature, to elucidate the interplay between magnetism and superconductivity.

We are interested in the $(\frac{1}{2}\frac{1}{2}1)$ antiferromagnetic reflection[1]. Although weak, is the strongest magnetic reflection.

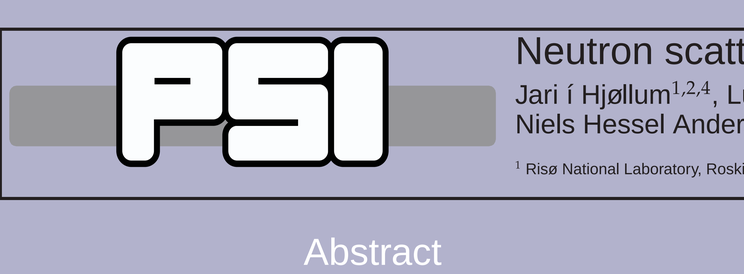

Fig. 1: **Left:** *A schematic of the YBCO unit cell, and a schematic of an YBCO nanoparticle.* **Center:** *TEM image of an YBCO nanoparticle. The bar is 10 nm.* **Right:** *TEM image of an YBCO nanoparticle. The bar is 50 nm.*

Sample preparation

The YBCO nanoparticles were prepared by a citrate gel modification of the sol-gel technique as described in [2, 3], and subsequently reduced in a N atmosphere to zero oxygen doping. The mean size of the YBCO nano-particles have been determined by Rietveld refinement of x-ray diffraction to approximately 40 nm in diameter and to be about 3-10 nm thick.

The particles have been stripped of water and oxygenated in a gas volumetric system. The water is removed by slowly (over the course of a day) raising the temperature of the sample to 600°C, and removing the water vapor by a cold trap (liquid N_2).

The doping is set by regulating the equilibrium pressure and temperature according to the phase diagram [5, 6], and allowing the sample to absorb the required amount of oxygen. Afterwards, the sample is sealed and cooled during a few hours. We have so far prepared two samples (sample 1 and 2) which both originally are parts of the same YBCO nano-particle powder. These have dopings $x = 0.2 \pm 0.025$ and $x = 0.3 \pm 0.025$ (estimated uncertainties). Each sample contained approximately 2 g of material.

Experiment

The samples have been investigated using the triple axis spectrometer RITA-II at the SINQ neutron source, Paul Scherrer Institute in Villigen, Switzerland.

The RITA-II spectrometer has 7 analyzer blades and a position sensitive detector(PSD), and can therefore measure 7 data points simultaneously (see. fig. 2. (right)).

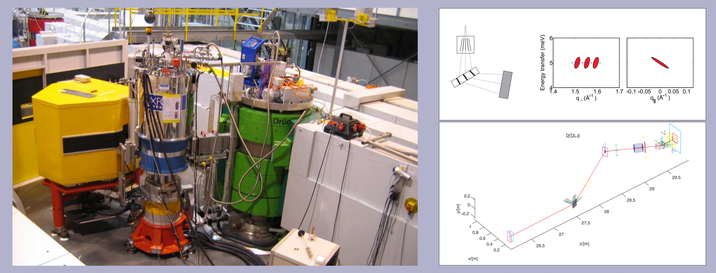

Fig. 2: **Left:** *Image of the RITA-II spectrometer. From the left, the yellow detector tank containing the analyzer assembly and the PSD. The sample is located further right, inside the blue cryo magnet. The monochromator is located inside the green tank. The grey box to the right of the monochromator tank contains the neutron guide.* **Top-right:** *A schematic of the RITA-II analyzer setup. Also shown is a resolution function for a multi blade mode (3 blades).* **Bottom-right:** *A schematic of RITA-II.*

Different arrangements of the analyzer blades, assembly and PSD, give rise to a variety of different user modes. The mode that we have used is the **monochromatic imaging mode** where neutrons are reflected onto separate regions on the PSD. In this mode, the setting of the blades is such that they all scatter neutrons of the same energy. As can be seen from fig. 2 the analyzer blades collect different scattering angles. Hence different q's are detected on different regions on the PSD. By defining software windows (counting bins) appropriately, the RITA-II acts as 7 triple axis spectrometers working in parallel. The data from the individual windows can later be combined, which henceforth can be analyzed in a standard fashion. Examples of combined spectra are presented in fig. 3.

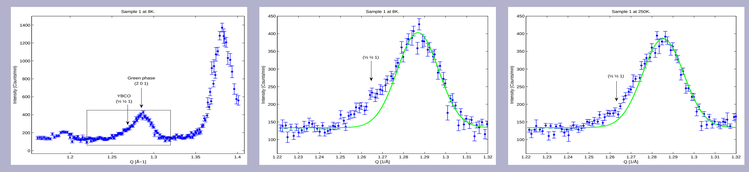

Fig. 3: **Left:** *A raw, combined spectrum of sample 1 at T=8 K* **Center:** *A zoom on the raw,combined spectrum of sample 1 at at T=8 K, on the $(\frac{1}{2}\frac{1}{2}1)$ (marked) antiferromagnetic reflection in YBCO at $q = 1.26$ Å^{-1}. The green gaussian represents the "green phase" peak.* **Right:** *A zoom on the raw, combined spectrum of sample 1 at at T=250 K.*

In our experiments we have investigated the $(\frac{1}{2}\frac{1}{2}1)$ antiferromagnetic reflection in the YBCO nanopowder at different T (8-450 K) in zero field by powder diffraction.

In addition to the YBCO reflections the spectra contain structural reflections from the non-magnetic impurity $Y_2Cu_2O_5$, the so-called "green phase". Namely the phase $(2\,0\,1)$ reflection which is located at q=1,268 Å^{-1}, may be disturbing since it is located adjacent to the $(\frac{1}{2}\frac{1}{2}1)$ magnetic reflection of YBCO.

Fitting Strategy

In stead of describing the fitting procedure in detail, we provide the most important factors in the fitting work.

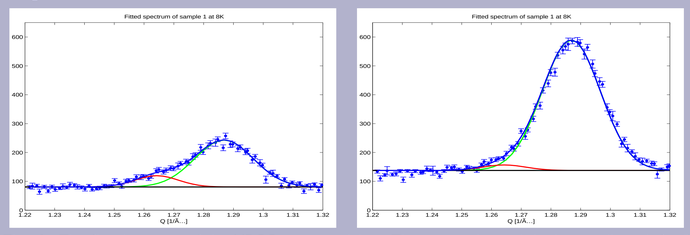

Fig. 4: *The fitted spectra of the YBCO nanoparticles. The magnetic peak is the small peak (red) on the left slope of the big peak (Green phase)(green). Notice the difference of the intensity of the magnetic peak. The spectra have been scaled to a comparable scale.* **Left:** *Doping x=0.2 at T=8 K.* **Right:** *Doping x=0.3 at T=6 K.*

- All fitting performed on a cutout of the full spectrum.
- The central peak fitted with two gaussians, one for the "green phase" structural peak and one for the magnetic peak.
- All fit parameters free at lowest temperature.
- Only amplitude of peaks and background allowed to vary freely at all other temperatures.
- Width of peaks constant.
- Position of peaks shifted linearly, according to $q(T) = \Delta q \cdot T + q_0$.
- $\frac{\Delta q}{q_0}$ is equal to the same constant for all peaks.

Discussion

The normalized magnetization curve obtained for the two dopings has been fitted to the formula $M(T) = M\left(\frac{(T_N - T)}{T_N}\right)^{\beta} + M_0$, and the result is presented in fig. 4. We have fitted three models: A bulk model (mean field theory) where β has been kept fixed at 0.5. A Model in which T_N is fixed according to data from [1], and a free model in which all parameters have been allowed to vary.

Fit	M	T_N(K)	β	M_0	χ^2
Fixed β	15.6	390	**0.50**	6.71	1.54
Fixed T_N	14.8	**390**	0.61	7.90	1.44
All free	15.7	400	0.59	7.05	1.99

Fit	M	T_N(K)	β	χ^2
Fixed β	14.9	461	**0.50**	0.175
Fixed T_N	13.7	**360**	0.16	0.110
All free	13.4	350	0.12	0.161

Left: Tab. 1: *Doping x=0.2.* **Right: Tab. 2:** *Doping x=0.3.*

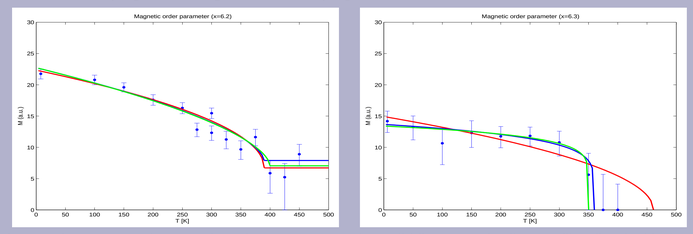

Fig. 6: *The measured magnetic order parameter. The solid lines are the line profiles fitted with the data.* ***1)*** *The red curve is the fitted magnetic order parameter as a function of temperature as predicted by mean field theory ($\beta = 0.5$).* ***2)*** *The blue curve is the fitted magnetic order parameter fitted with a fixed T_N.* ***3)*** *The green curve is the fitted magnetic order parameter freely fitted.* **Left:** *Doping x=0.2.* **Right:** *Doping x=0.3.*

We have proven the existence of the antiferromagnetic phase in nano-sized YBCO particles.

The Néel temperature decreases with increasing oxygen doping from 400 K to 350 K for x=0.2 and x=0.3 respectively. The β parameter decreases significantly from 0.59 to 0.12, which may be related to the nano-size of the YBCO particles. And finally M(0) decreases from 15.7 to 13.4. For this nanoparticle size the Néel temperatures correspond to the bulk values.

In our future work we will focus on studies of the Néel temperature and mapping of the superconducting phase transition with decreasing particle diameter. Furthermore we will acquire data with an applied field.

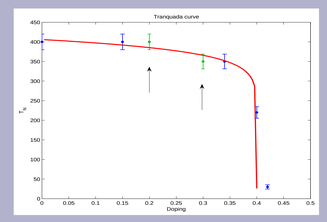

Fig. 6: *Data from [1] combined with our data. Our points are green and marked with arrows.*

References

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