Experimental report

17/09/2018

Proposal: **4-02-486**

Council: 4/2016

Title: Antiferromagnetic excitations in the very underdoped model cuprate HgBa2CuO4+x

Research area: Physics

This proposal is a new proposal

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Samples: HgBa2CuO4+x

Instrument	Requested days	Allocated days	From	To
IN8	10	7	19/06/2018	26/06/2018

Abstract:

The cuprates exhibit strong antiferromagnetic (AF) fluctuations even in the superconducting doping range. The prominence of these fluctuations has led to suggestions that they may be involved in the pairing necessary for high-temperature superconductivity. Hence extensive studies have been carried out in La2-xSrxCuO4 (LSCO) and YBa2Cu3O6+x (YBCO), which revealed an hourglass dispersion and related resonance characteristics. In our recent study of the structurally simple model cuprate HgBa2CuO4+x (Hg1201), we observed distinct new features, along with a strong doping dependence of these features. In order to better understand the role that AF fluctuations play in the cuprates, we propose to extend our study to a very underdoped sample with Tc less than 40K.

Antiferromagnetic excitations in the very underdoped model cuprate $HgBa_2CuO_{4+x}$

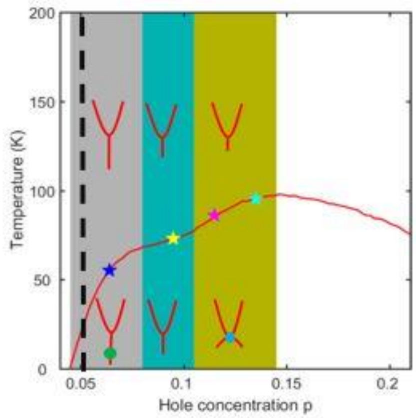

***Figure 1**: Hg1201 phase diagram showing the general shape of the AF dispersion as a function of doping and temperature.*

The high-temperature cuprate superconductors exhibit prominent antiferromagnetic (AF) correlations over a large region of the temperature-doping phase diagram, indicative of a link between the AF correlations and the superconducting and pseudogap states. Consequently, extensive theoretical and experimental work has been devoted to the study of the AF correlations in the cuprates. Neutron scattering measurements of $La_{2-x}Sr_xCuO_4$ (LSCO) and $YBa_2Cu_3O_{6+x}$ (YBCO), for which large single crystals have long been available, have found a seemingly universal 'X'- or hourglass-shaped magnetic dispersion centered at the two-dimensional AF wave vector ($\mathbf{q}_{AF}$ = (½, ½) r.l.u.). We have been investigating the AF response in $HgBa_2CuO_{4+\delta}$ (Hg1201), a simple tetragonal cuprate with a single copper-oxygen plane per unit cell, and the highest optimal superconducting transition temperature (T_c = 97 K) among all single-layer cuprates. Our previous neutron scattering studies of Hg1201 have shown that the AF response is 'Y'-shaped, in contrast to what is observed in other cuprates (Fig. 1). An 'X'-shape is recovered at large hole doping in the SC state only. Our prior work focused on samples between optimal doping and p ≈ 0.064 (corresponding to a T_c of 55 K) on the underdoped side of the superconducting dome. More heavily underdoped samples were not yet available.

The goal of this experiment was to obtain initial information about the AF response in a new sample with unprecedentedly low doping level of p ≈ 0.05 (T_c = 29 K, sample mass = 0.6 g). In particular, we wanted to obtain initial data to determine: (i) if a AF excitation gap is still present so close to the non-superconducting state; (ii) the energy dependence of the magnetic response; (iii) the temperature dependence of the response, particularly across T_c; (iv) and, importantly, whether the response is commensurate or incommensurate at low energy.

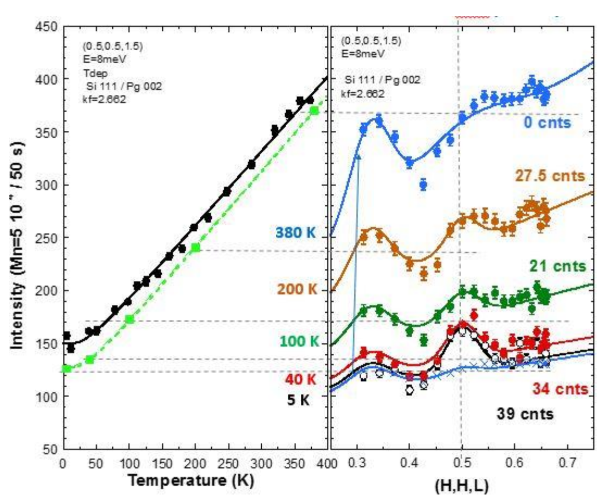

***Figure 2** Left: T-dependence at (½, ½, 1.5) and E=8 meV: scattered intensity (black), background from rocking scans (green). Right: corresponding rocking scans.*

The thermal TAS IN8 was set up with the Si111 monochromator, and the sample was oriented in the (110)/(001) scattering plane. At low doping, spin fluctuations are expected to persist up to high temperature. Therefore, the sample was mounted inside a cryo-furnace in order to perform measurements from 2 K up to 400 K. Air leakage at the top of the cryo-furnace strongly affected the quality of our measurements during the first few days, until the cause of the anomalously high background in data was finally identified. Since this experiment was the first measurement of such a heavily underdoped sample of Hg1201, a significant amount of time in the first half of the experiment was used to roughly measure the

AF response and determine good values of the out-of-plane momentum transfer *L* and energy transfer at which to measure with longer counting times.

These tests led us to focus on rocking scans. Figure 2 shows the temperature dependence of the scattered intensity at $\mathbf{Q}_{AF}$ = (½, ½, 1.5) and a set of rocking scans, performed around that wave vector, at various temperatures. A signal centered at $\mathbf{Q}_{AF}$ was observed at low temperature. This signal gradually vanishes with increasing temperature, as expected for AF spin fluctuations. The temperature dependence of the scattered intensity at $\mathbf{Q}_{AF}$ = (½, ½, 1.5) was measured (Fig. 3), in addition to the temperature dependence of the nuclear background on top of which the AF fluctuation develops in the rocking scans. After subtraction of this background and correction for the detailed-balance factor, we obtained the temperature-dependence of the imaginary part of the dynamical susceptibility, $\chi''(\mathbf{q}_{AF}$, 8meV) (Fig.3-A), which confirms the decrease of the AF response upon heating.

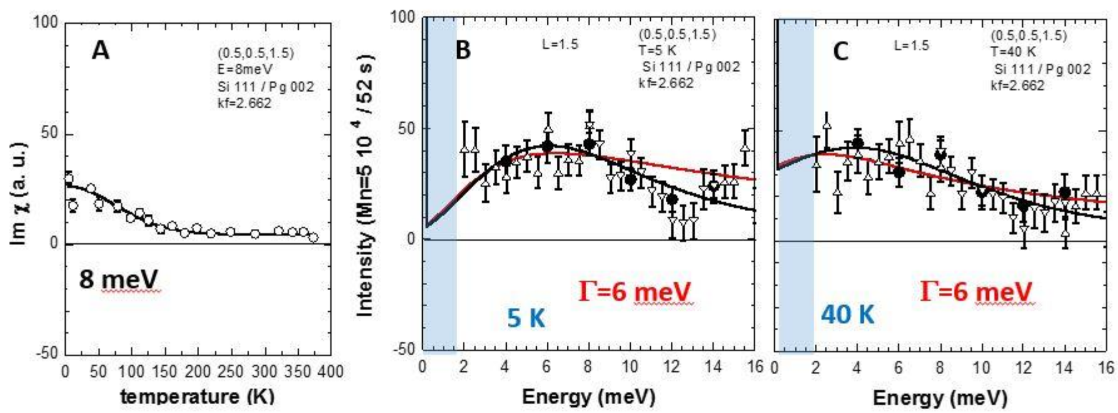

Figure 3 *(A) T-dependence of $\chi''(\mathbf{q}_{AF}$, 8meV). Energy dependence of S($\mathbf{q}_{AF}$,E) at 2 K in the (B) superconducting state and (C) at 40 K in the normal state. The black and red lines correspond to fits with damped harmonic oscillator and single relaxor functions, weighted by the detailed balance factor. Monitor corrections for the Si111 monochromator are neglected.*

Figures 3B-C show the energy dependence of the spin-spin correlation function S($\mathbf{q}_{AF}$,E), measured at 2 K in the superconducting state and 40 K in the normal state. Rocking scans at fixed energy and complementary E-scans on both side of the AF wave vector were used to extract the magnetic scattering from the raw data at $\mathbf{q}_{AF.}$ While previous studies highlighted the large spin gap (extremely robust even at high temperature), our new data for highly underdoped Hg1201 demonstrate that commensurate AF fluctuations are present at the lowest energy (3 meV). The analysis of S($\mathbf{q}_{AF}$,E) using a single relaxor or a damped harmonic oscillator suggests that a characteristic energy scale of about 6 meV. As can be seen from Fig.3B-C, the low-energy part of the spin excitation spectrum does not display a significant change across the superconducting transition. This insensitivity to the superconducting transition is often observed in lightly-doped cuprates, in particular when a glassy magnetic state tends to develop at low temperature.

The observation of low-energy AF fluctuations in a highly underdoped sample of simple tetragonal Hg1201 is the main result of our study. More work is need on a cold TAS to search for the existence of a glassy magnetic state in this sample that may co-exist or compete with the superconducting state at low temperature. Moreover, our study extended to high energy (~50 meV), and these initial data will also be used for comparison with future TOF measurements devoted to the high-energy dispersion of the spin excitation spectrum.