Experimental report

05/09/2023

Proposal: **4-02-620** **Council:** 10/2022

Title: Antiferromagnetic response of the extremely underdoped model cuprate HgBa2CuO4+d

Research area: Physics

This proposal is a new proposal

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Samples: HgBa2CuO4+d

Instrument	Requested days	Allocated days	From	To
IN8	10	7	15/05/2023	22/05/2023

Abstract:

The undoped cuprates are antiferromagnetic (AF) Mott insulators which produce a range of complex electronic and magnetic phenomena when doped with holes, including high-temperature superconductivity, T-linear resistivity in the normal state, charge density wave order, and the opening of a pseudogap at the Fermi level. Much of the physics of the normal state and the pairing mechanism that results in superconductivity upon cooling remains contested, though several theoretical descriptions indicate that AF correlations play a central role. In many cuprates, notably La2-xSrxCuO4 and YBa2Cu3O6+d, these correlations consist of magnetic excitations with an X-shaped dispersion centered on the AF wave vector. These materials are complicated by low structural symmetry or and/or disorder which mask the underlying physics of the CuO2 planes. In previous measurements of moderately underdoped HgBa2CuO4+d (Hg1201) - a simple-tetragonal compound - we found a Y-shaped AF dispersion indicating the response of pristine CuO2 planes. We propose to continue these measurements with a new extremely-underdoped sample, near the edge of the superconducting dome, and with an AF gap of only a few meV.

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Title: Antiferromagnetic response of the extremely underdoped model cuprate $HgBa_2CuO_{4+\delta}$

Experimental Team

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Environments: Cryo-furnace

Experimental report:

The IN8 experiment was carried out on an array of $HgBa_2CuO_4$ (Hg1201) single crystals of total mass ~1 g, co-aligned on Al plates. The full mosaic of the sample is about 1.8°. The overall superconducting transition temperature obtained from individual measurements of each crystal is ~32 K: this highly underdoped Hg1201 sample (UD32) is lightly hole doped, with an estimated hole doping of ~5% / Cu. We used Si111 as monochromator and Pg002 as analyzer. Both monochromator and analyzer were focused in order to the increase the neutron flux on the sample (height 2.5 cm, width 1 cm). A PG filter was installed on the scattered neutron beam to remove higher-order contamination. The final neutron wave vector was set to either k_f = 2.661 A-1 or 1.97 Å^{-1}. The sample was installed within a cryo-furnace (maximum temperature of 550 K) and aligned in the (100)/(010) plane, so that wavevectors of the form (H K 0) were accessible. Neutron data are reported in reduced lattice units (r.l.u) (2π/a 2π/a 0).

Prior to the experiment, the spin excitation spectrum was studied using ToF measurements on ARCS at SNS. The imaginary part of the dynamical magnetic susceptibility at base temperature and at 45 K (~13 K above T_c) showed spin fluctuations centered at the AF magnetic wave vector, $\mathbf{Q}_{AF}$ = (0.5 0.5 0), up to about 50 meV, and dispersing spin fluctuations at higher energies. The low-energy spin fluctuations were maximum around 10-12 meV. Following this ToF study, we focused our attention on three open issues: Is there a change in the response on passing through T_c? Are the low-energy spin fluctuations truly commensurate? Is there a spin gap?

The IN8 experiment was carried out at T = 1.5 K (<< T_c) and 45 K (> T_c). E-scan and constant E-scans were combined to characterize the magnetic signal around $\mathbf{Q}_{AF}$ that appears above a smooth nuclear background. The study was performed around (0.5, 0.5,0) in the E-range 1-11 meV at k_f = 1.97 Å^{-1}, 2-14 meV at k_f = 2.661 Å^{-1}, whereas around (1.5,0.5,0), the E-range was 2-40 meV at k_f = 2.661 Å^{-1}. After nuclear background subtraction and correction for monitor and the detailed-balance factor, the imaginary part of the dynamical magnetic susceptibility, Imχ($\mathbf{Q}_{AF}$,E), was obtained using the square of the Cu-magnetic form factor.

The magnetic spectra at 1.5 K and 45 K, up to 40 meV (Fig.1-A), are consistent with the ToF data. The main change of intensity on warming above T_c is below 15 meV. At 1.5 K, the magnetic response is not discernible anymore at 1 meV (Fig. 1-B), suggesting the existence of a true spin gap between 1 and 2 meV. The data at higher energy display a gaussian profile (Fig.1B). In other lightly hole-doped single layer cuprates (e.g., $La_{2-x}Sr_xCuO_4$), the magnetic response is incommensurate around p = 5% holes/Cu and the magnetic scattering is made of a quartet of peaks at $\mathbf{Q}_{AF} \pm (\delta',\delta')$ and $\mathbf{Q}_{AF} \pm (\delta',\delta')$, with $\sqrt{2}\delta' = \delta = p$. In Hg 1201 UD32, one cannot rule out the existence of such incommensurate spin fluctuations, but such incommensurate fluctuations would need to be extremely broad, with a typical FWHM of ~δ. This would imply that the magnetic correlation length is comparable to the pitch of the modulated magnetic scattering.

Figure 2.A shows the temperature dependences of the signal measured at E={1,2,9} meV, where the nuclear background is extracted from rocking scans at fixed temperatures. Fig. 2B-C show the T-dependences of the magnetic scattering at 9 and 1.5 meV. At 9 meV, the magnetic scattering decreases continuously upon warming. A change at T_c is consistent with our data. At 1 meV, on the other hand, the T-dependence is more complex and approximately follows the form 1/T exp($-\Delta/k_BT$) with Δ = 0.4 meV. If such a T-dependence were confirmed in a cold triple-axis experiment, this would imply the existence of a second energy scale at low temperature, possibly due to an evolution of the superconducting order

parameter from pure d-wave symmetry to s+d symmetry. More work is needed to clarify that aspect using a cold neutron beam.

We made complementary (quasi-)elastic measurements in order to search for the existence of a (quasi-)static AF response, coexisting with the superconducting order. Unfortunately, the higher order contamination (λ/4) was too larger to prevent a full study of a (quasi-)elastic AF response. Similar to the low-energy dynamic response, the question of the existence of (quasi-)static AF correlations should be addressed using a cold neutron beam, as a continuation of the IN8 experiment.

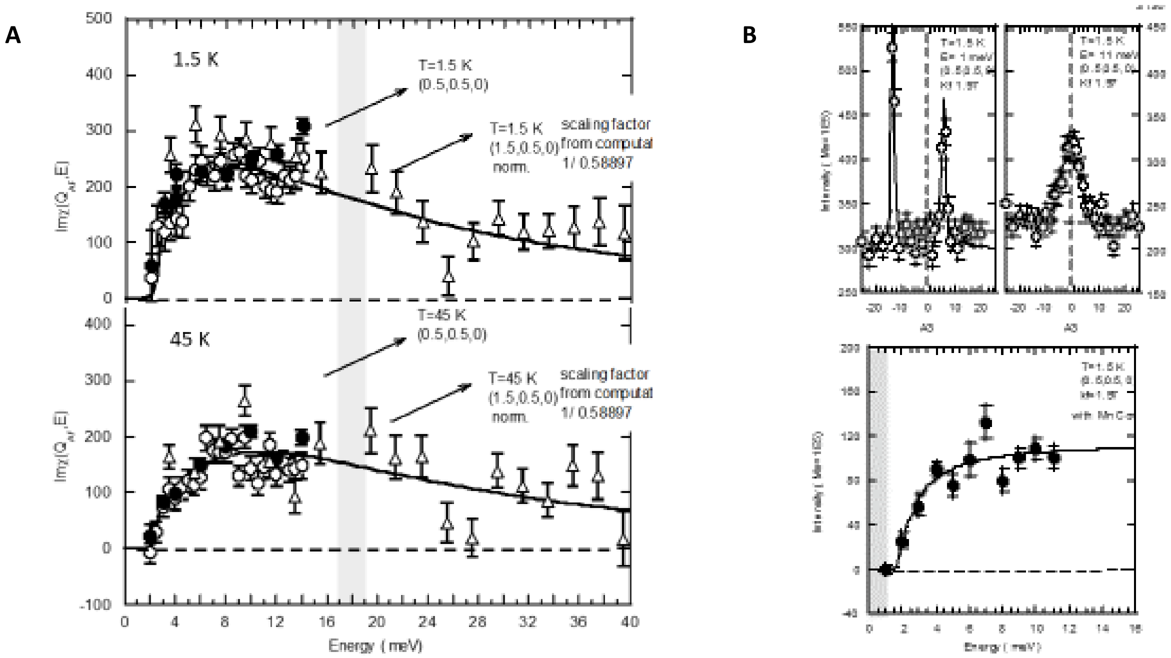

*Fig. 1 A, Imaginary part of the dynamical magnetic susceptibility, Imχ(**Q**$_{AF}$,E), measured at 1.5 K (top) and 45 K (bottom) with k_f = 2.661 Å^{-1} . **B**, Imχ(**Q**$_{AF}$,E) obtained from rocking scans at fixed energy at T = 1.5 K with k_f = 2.661 Å^{-1} . Top left: rocking scan at 1.5 K with no discernible AF response at 1 meV. Top right: rocking scan with a net AF response at 11 meV. The latter signal is well centered at **Q**$_{AF}$. A Gaussian fit gives a full width at half maximum (FWHM) of 0.09 r.l.u, much larger that the experimental resolution of 0.02 r.l.u.*

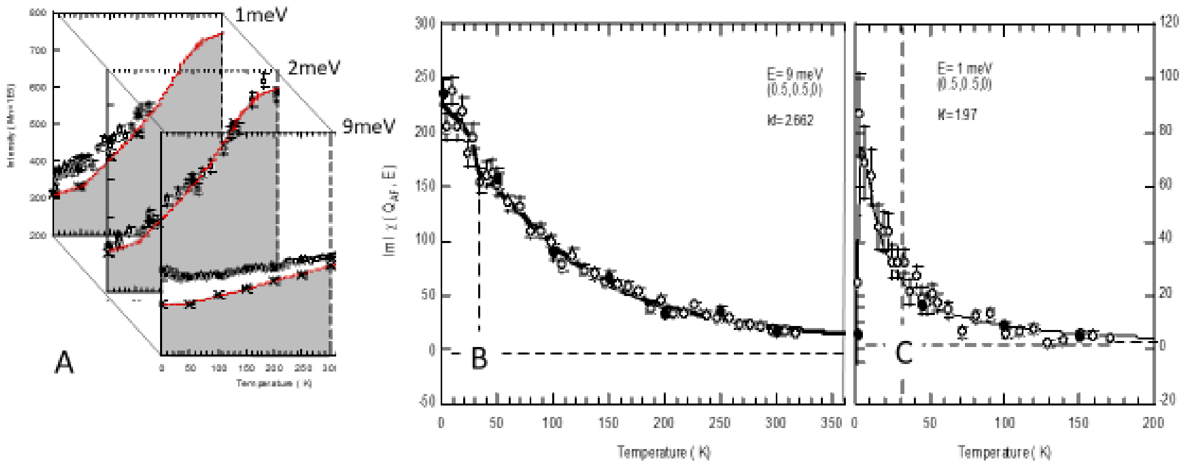

***Fig. 2** A: T-dependences of the scattered intensity at {1, 2, 9}meV, where crosses correspond to the nuclear background deduced from rocking scans. B-C) T-dependences of the imaginary part of the dynamical susceptibility at Q_{AF} measured at 9 meV (k_f = 2.661 Å^{-1}) and 9 meV (k_f = 1.9 Å^{-1}). The respective intensities differ by a factor of 2.45, which corresponds to the expected k_f^3cotan(θ_A) change in the instrumental resolution.*