	Experiment title: Looking for pair density waves in $YBa_2Cu_3O_{7-\delta}$ via RIXS	**Experiment number**: HC4607
Beamline: ID32	**Date of experiment**: from: 18/01/2022 to: 24/01/2022	**Date of report**: 08/03/2022
Shifts: 18	**Local contact(s)**: Dr. Nicholas Brookes	*Received at ESRF:*

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Report:

The doping-temperature phase diagram of cuprate high critical temperature superconductors (HTS) is populated by several symmetry-breaking orders, whose intertwining and competition is considered the key to solve the grand unresolved issues of this class of materials. Some of these orders, as spin density waves (SDW) and charge density waves (CDW) have been disclosed and investigated by many experiments performed in the last two decades. Others instead, even though have been theoretically predicted, still remains inaccessible or experimentally elusive. This is the case of as the pair density wave (PDW) order, a new state of matter with unique properties not encountered in other superconductors. Such emerging order should be characterized by Coopers pairs having a finite momentum Q and therefore generating states with spatially modulating Cooper pair density [1]. Theoretically, *the PDW order would represent a sort of "mother state" for other HTS orders, inducing both the charge and the nematic orders*.

Up to now the evidence for the existence of PDW came first, indirectly, by transport experiments on 214 cuprates at doping p=1/8 [1]; more recently, by STM experiments on underdoped $Bi_2Sr_2CaCu_2O_{8+x}$ [2,3], where a Cooper-pair density modulation has been directly visualized. These experimental observations of PDW are still widely debated. The reason is mainly twofold: on one side, PDW has been observed only in limited areas of the cuprate phase diagram, and in few cuprate families. On the other side, PDW modulations should also appear with a wavelength about twice that of CDW, i.e. $\lambda_{PDW}\approx 2\lambda_{CDW}\approx 8a_0$. However, such CDW subharmonic with a wavevector $q\approx q_{CDW}/2$, has been observed only by STM, which is strongly surface-sensitive, but never by X-ray scattering experiments, which have offered over years the strongest evidence of CDW in cuprate HTS.

A recent test we have performed at ID32 on a strongly underdoped (T_c^0 = 12 K) $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin film gave us an indication of the presence of PDW and of its detectability with X-rays. By integrating the quasi elastic region of the RIXS spectra measured along the (H,0) direction, we disclosed both a charge order peak at $q_{CDW}\approx 0.32$ rlu and a second peak at c. We have achieved this, by properly subtracting the intensity coming from the specular centered at Γ=(0,0), which covers any small signal below q=0.2 rlu.

Based on the aforementioned test, we have decided to investigate, in the framework of the experiment HC4607, this second peak at low momentum value q^{2nd} in $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films covering a broad range of doping. The usage of several doping levels is crucial to determine the doping range where the second peak is present: this can be crucial to understand the pairing mechanism of PDW and its intertwining with the charge order phenomenon.

We have performed Cu-L3 RIXS measurements on three superconducting YBCO samples, grown at Chalmers University [4], with doping p=0.06 (no CDW and weak charge density fluctuations - CDF), 0.12 (strongest CDW), and 0.19 (no CDW and strong CDF). As a reference, we have also measured a $CaCuO_2$ (CCO) film, which is representative of a perfectly undoped cuprate.

As a first step, for each sample we have investigated the presence of a peak at $q_{CDW}/2$ in the quasi elastic region of the spectra (energy range between -40 meV and 100 meV), measured at several momentum values $q_{//}$ along the (*H*,0) direction of the Brillouin zone. The background signal, used to properly remove the specular intensity, has instead determined measuring the spectra along the (*H*,*H*) direction, and integrating the quasi-elastic region. The measurements have been performed at two/three temperatures between 20 and 260 K. A summary of the results we have obtained at 20 K in presented in Figure 1.

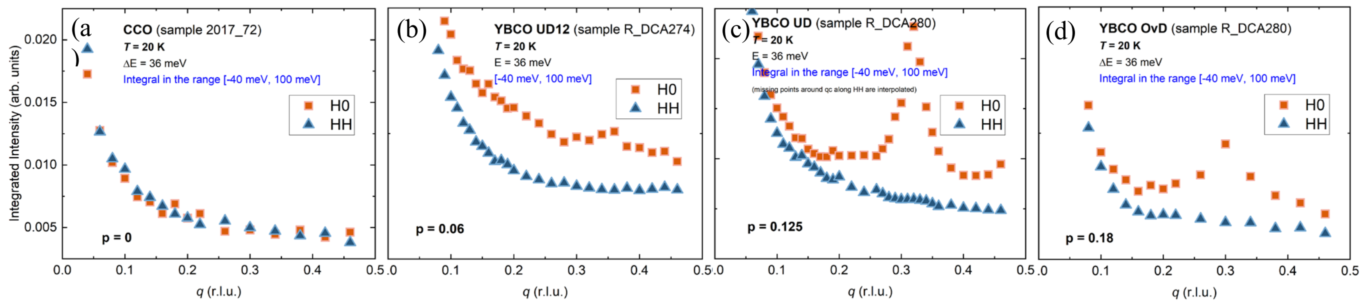

Fig. 1: Quasi-elastic intensity of the RIXS spectra measured along the (H,0) and (H,H) direction for (a) an undoped (p=0) CCO sample; (b) a strongly underdoped (p=0.06) YBCO sample; (c) an underdoped (p=0.125) YBCO sample and (d) a slightly overdoped (p=0.19) YBCO sample.

On the undoped CCO sample (Fig.1(a)), the quasi-elastic intensity measured at a fixed *q* is the same along the (*H*,0) and (*H*,*H*) directions. The two scans are instead profoundly different, in intensity and shape, in the three superconducting YBCO films (Fig.1(b)-(c)-(d)): at $q \approx 0.30$-0.35 rlu (depending on the doping) we clearly distinguish the charge order (CDW, CDF or both) peak along the (*H*,0) direction. In particular the broad CDF peak is also responsible of the intensity gap between the (*H*,0) and (*H*,*H*) directions which is visible at high *q* values [5]. Moving to small *q* values, in particular to $q \approx q_{CDW}/2$, the intensity of the (*H*,0) and (*H*,*H*) scans is rather similar for the two films at p=0.12 and p=0.19; viceversa, it is much stronger along the (*H*,0) direction for the strongly underdoped sample (Fig.1(d)). This might be the signature of a PDW signal. Moreover, this increased intensity is only present in YBCO at the p=0.06 doping, where SDW have been measured by neutron scattering at $q \approx 0.03$ rlu [6]. This doping dependence follows the line of the experimental evidence in 214 cuprates, where PDW looks strongly entangled to the SDW and CDW orders [7].

Based on the abovementioned result, we have performed on the strongly underdoped YBCO an azimuthal scan in the range between φ = 0 (*H0* direction) and φ = 45 (*HH* direction) at two different values of momentum: $q = 0.15 \approx q^{2nd}$, where we have indication of a putative PDW signal, and at $q = 0.44$, i.e. far from both the PDW peak and the CDF peak. We expect indeed that the presence of many entangled, symmetry-breaking orders in this sample may be at the origin of a (multi)-folded Brillouin zone. Consequently, that in the chosen azimuthal range the intensity of the collective excitations to which we have access in the RIXS spectra might present a series of discontinuities. While the quasi-elastic intensity changes rather smoothly as a function of φ, the intensity of the *dd* excitations (determining integrating the spectra in the range between -3 and -1 eV) undergoes unexpected jumps, superimposed to a smoother behavior (Fig. 2). This is quite surprising, since *dd* excitations are often dismissed as uninteresting because they have atomic nature and thus do not encode the relevant physics of cuprates. This preliminary result, which requires further investigation in the future, seems to confirm another recent result, again pointing to the precence of band-like effects occurring in the *dd* excitations of cuprates [8].

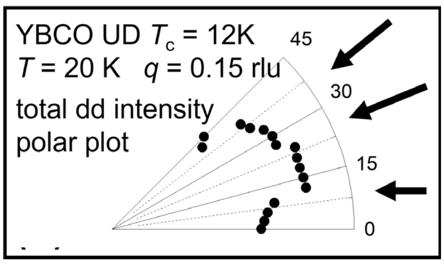

Fig. 2: The azimuthal scan, performed on a strongly underdoped YBCO film at q=0.15 rlu, shows several jumps of the *dd* intensity, highlighted by arrows.

The results presented in Figures 2 are preliminary and will require additional beamtime to be devoted specifically to this issue. The results based on the (*H*,0) and (*H*,*H*) scan, partially summarized only at one temperature in Figure 1, are instead rather solid but their interpretation will require a careful analysis. In general, we are looking to relatively small peaks/modulations in a range of wavevectors which is dominated by the huge intensity coming from the specular centered at Γ=(0,0). However, although the experiment has been extremely challenging from the technical point of view, the dataset looks extremely promising and the beamtime can be considered really successful. The final data analysis will be carried out with the greatest care with the aim of extracting the maximum of information.

This very first experiment confirmed the actual existence of the $q_{CDW}/2$ peak in YBCO. The dataset we have collected, and in particular the doping dependence we have determined, will provide to us, after careful analysis, indications on the actual PDW nature and on its relation to the CDW order.

References:

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