Comment on ”No Evidence for Orbital Loop Currents in Charge Ordered YBa$_2$Cu$_3$O$_{6+x}$ from Polarized Neutron Diffraction”

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Intra-unit cell magnetic order has been observed in four different families of high-temperature superconductors from polarized neutron diffraction experiments and supported by several other techniques. That order, which does not break translation symmetry, is consistent with the predicted spontaneous orbital moments generated by two microscopic loop currents in each CuO$_2$ cell. In a recent report, Croft et al [arXiv:1709.06128v1 claimed to find no evidence for such orbital loop currents in charge ordered YBa$_2$Cu$_3$O$_{6+x}$ using detwinned samples at least 100 smaller than in our experiments. We show by a detailed quantitative analysis of their data that contrary to their conclusion, the observed magnetic signal falls below the threshold of detection for their experiment. This is shown by noting that Croft et al overestimate by about an order of magnitude the expected magnetic signal. A factor ~ 4-5 is due to improper comparison of samples doping level, not taking into account detwinning of the samples and incorrect data calibration plus another factor of 3 in not accounting for possible finite correlation length. Not determining the spin-flip reference line properly, not doing a polarization analysis and inadequate control of the flipping ratio of the neutron beam add to uncertainties in their measurements.

I. INTRODUCTION

In an extensive series of papers [1–10], we demonstrated using polarized neutron diffraction (PND) that the pseudogap state of underdoped cuprate superconductors is characterized by a Q=0 magnetic order, also referred to as an intra unit cell (IUC) magnetic order [11]. That encompasses results in four different cuprates families with a large variety of dopings: YBa$_2$Cu$_3$O$_{6+x}$ (YBCO) [1–5], HgBa$_2$CuO$_{2+x}$ [6, 7], La$_2-\delta$Sr$_\delta$CuO$_4$ [8] and Bi$_2$Sr$_2$CaCuO$_{8+\delta}$ [9, 10]. Two reviews were written to give more experimental and technical details and put the different neutron results in perspective with the other physical properties of high-temperature cuprates [11 12]. In a recent manuscript posted on the arXiv, Croft et al [13] claimed that they ”found no evidence for the appearance of magnetic order below 300 K” in two YBa$_2$Cu$_3$O$_{6+x}$ samples.

We here show that Croft et al [13] could not observe the magnetic signal owing to the insufficient detection capability of their measurements. This is due to multiple limitations of their data and analysis, that are detailed below, spanning incorrect sample comparison, ignorance of the impact of detwinning, misleading data calibration and improper data collection strategy. We describe each of these shortcomings below and show that their estimation of the magnetic signal reported by us is wrong by an order of magnitude. Therefore, it is below the sensitivity of the experimental results of Croft et al [13]. The claimed upper bound for a possible magnetic moment is therefore not correct and should be disregarded. Further, the comparison with local probes results in [13] is oversimplified as it overlooks all the recent literature about muon spin resonance results [14, 15] leading to the possibility of finite time-scales of the IUC order. Such short range correlations have been actually reported in a recent study in nearly optimally doped YBCO [4] using PND.

The polarized neutron diffraction experiment we discuss here is very challenging [11]. It is worth emphasizing that neutron polarization analysis has been employed to prove the magnetic nature of the reported signal [1–4, 6–10]. The polarization sum-rule is systematically satisfied. We emphasize as well than the YBCO data are nearly indistinguishable from those for HgBa$_2$CuO$_{4+\delta}$ [6, 7]. There is no doubt left about the existence of a magnetic signal.

Over the years, the data analysis has been improved (see for instance, the refine analysis on the sample C from our first report in Fauqué et al [1] compared to the recent one in Mangin-Thro et al [5]). The earlier results [1] have been upgraded for quantitative accuracy: it reveals for the first time the IUC magnetic signal in five different YBCO samples and its striking evolution with hole doping, following the pseudogap physics. Gradually, the data analysis was more quantitative in the subsequent publications in YBCO [2, 5]. This has important consequences on the magnetic signal amplitude and the confidence one can get from it. The neutron intensity is proportional to sample mass. By using samples ~ 100 times smaller than ours on a spectrometer with about 3 times larger neutron flux (at the used wavelength), Croft et al [13] are thus facing more than 10 times more experimental issues. Even after a counting time of a few hours, their experiment
does not reach the required sensitivity.

We consider below different aspects which affect noticeably the comparison made by Croft et al.\cite{13} with our data and analysis. We demonstrate that various mistakes invalidate their conclusion and that they did not have enough sensitivity to detect the IUC magnetic signal in their measurements. In section II, we first recall a few key results, already present in the literature, that, put together, show that Croft et al.\cite{13} overestimated by about a factor $\sim 4$-5 the amplitude of the signal they were looking for. In section II we compare the data collection strategy in the studies carried out by Fauqué et al on the one hand and Croft et al. on the other hand. This comparison shows that Croft et al.\cite{13} overlooked important experimental features discussed previously\cite{1},\cite{11}, that limit severely their accuracy. Next, in section II, we underline the recent evolution of muon spin resonance ($\mu$SR) data which suggests that the magnetic correlations are at short range with important consequences for the detection limit in PND, since instrumental resolution effects have to be considered very carefully. Finally, in section II we present an improved analysis of our original paper Fauqué et al.\cite{1} by taking account of the instrument resolution, leading only to a small change, about 15% in the conclusion which were already reached. We can then assess a direct comparison with the data reported by Croft et al.\cite{13} and demonstrate how the IUC magnetic signal falls below the threshold of detection of their measurements.

II. DOPING LEVEL AND NEUTRON STRUCTURE FACTOR

A. Sample comparison

The table\cite{1} shows all the underdoped YBCO samples utilized in PND experiments. Croft et al.\cite{13} focussed exclusively on the pioneering work of Fauqué et al.\cite{1} neglecting further results. Their comparison with the previous results is also questionable as they compare samples which are characterized by distinct hole doping levels.

In four independent measurements on three different underdoped samples with superconducting transitions within the so-called 60K-plateau (samples YBCO$_{6.6}$), very reproducible results were found: that corresponds to sample B and C for\cite{1,2} and the report of Mook et al\cite{2} (sample labelled here M) on a different sample. Two of these samples (samples B and M) are twinned and exhibit an oxygen ordering, ortho-II, corresponding to one Cu-O chain filled over two (see e.g. ref.\cite{18} for an extensive discussion of the oxygen ordering in YBCO). The third sample is detwinned and has a different ordering of extra oxygen in CuO-chains, ortho-VIII\cite{18,20}. For all these reports, the magnetic cross-section at the Bragg position $Q=(011)$ is systematically found to be $1.6 \pm 0.1$ mbarn (see table\cite{1}). That is the correct order of magnitude which should be used to carry out a comparison with our data. The extrapolation of ”Fauqué et al” in Figures 8 and 11 (for sample H1) of Croft et al is then overestimated by a factor 2 by this first issue. As sample H1, both samples B and M show the ortho-II oxygen ordering whereas sample C exhibits the ortho-VIII oxygen ordering as sample H2.

B. Impact of detwinning

Next, both samples studied by Croft et al.\cite{13} are detwinned samples. However, the possible impact of detwinning on the neutron structure factor is ignored. Even if the IUC structure factor is not expected to change upon an orthorhombic distortion, it was experimentally proved that it had an important impact in YBCO on the neutron structure factor of the IUC order\cite{2}. Indeed, that study of detwinned YBCO reveals a L-dependent a-b anisotropy of the scattered magnetic intensity, pointing out that the bilayer mirror plane is lost. Such an effect can be accounted for by a stacking of criss-crossed magnetic pattern within a bilayer. For $L=0$, the magnetic intensity is predominantly along $a^*$ and weaker along $b^*$. However, that difference between $a^*$ and $b^*$ is negligible for $L=1$. In terms of loop current models\cite{18,17}, it means that the sum of the toroidal moments of each plane of the CuO$_2$ bilayer is pointing along $b^*$, the direction of the Cu-O chains\cite{2}.

That study was made in the ortho-VIII (sample C), the same can be expected in ortho-II sample as what matters is the locking of the composite toroidal moment along the...
CuO chains, b*. The magnetic scattering at the (010) Bragg reflection appears \(\sim\) 3 times weaker than that at the (100) reflection \([3]\). Croft \textit{et al} \([13]\) ignores that result and compares their result on the (010) reflection with our sample A which was twinned where the magnetic intensity is predominantly controlled by the (100) reflection. As a result, an intensity of 2.0 mbarn at 70 K for the (010) reflection is expected for a detwinned sample instead of 9.0 mbarn (expected from twinned sample A) as is misleadingly reported in Fig 11.a of Ref. \([13]\). This has a strong impact on the figure 8.a and figure 11.a of Ref. \([13]\). The expected amplitude from Fauqué \textit{et al} \([1]\) for a detwinned sample with an ortho-II structure is compatible within the statistical accuracy of the shown data for the (010) reflection (see below section \(\bigstar\) and Fig. \(\bigstar\)).

### C. Data calibration

A third issue in the report of Croft \textit{et al} \([13]\) is related to the use of a procedure of calibration of the data in absolute unit which differs substantially from the one systematically used in previous PND studies \([1,2]\). As explained in the figure 1 caption of ref. \([1]\) and in \([11]\), the magnetic cross sections were calibrated in mbarn using the nuclear Bragg cross section of the (004) nuclear Bragg reflection. One then needs to estimate the nuclear neutron structure factor \(F_N\) which reads:

\[
F_N = \sum_n b_n e^{i\mathbf{Q} \cdot \mathbf{R}_n},
\]

where \(b_n\) and \(\mathbf{R}_n\) correspond to the neutron scattering length and the position of the n-th atom in the unit cell. A calculation of that nuclear structure factor gives \(f_{004}^{\text{calc}} = |F_N|^2 = 7\) barn \([1]\) using the measured atomic positions \([22]\). The Bragg peak (004) was chosen as we had to compare intensity of 5 different (twinned and detwinned) samples all together with different oxygen content. As discussed below, the calculated structure factors, \(|F_N|^2\), depend on the specific chain-oxygen ordering. However, this effect is limited for the (004) reflection; the estimate of 7 barns is an averaged value over the doping range given by table \([\bigstar]\). Actually, a collection of Bragg peaks intensity along (00L) direction has been measured and can be used to calibrate the magnetic intensity of our samples (see section \(\bigstar\) and Fig. \(\bigstar\) below). The Bragg peaks cross-sections along (00L) are typically less sensitive to specific oxygen ordering of oxygen atoms in the Cu-O chains.

Next, we use the intensity ratio (or cross-section ratio) of the nuclear Bragg peak to magnetic one \(|F_N/F_{\text{mag}}|^2\). As detailed in \([11]\), we found the smaller ratio \(|F_N/F_{\text{mag}}|^2 = 400\) at 70K for the (011) reflection of our detwinned sample C. That gives \(|F_N|^2 \sim 0.5\) barn for the (011) nuclear cross-section, about two times larger than the value quoted by Croft \textit{et al} \([13]\) of 0.28 barn. That leads to an additional overestimation by a factor 2 of the magnetic intensity of Fauqué \textit{et al} \([1]\) in figures 8.e, 9.c and 11.b,c Croft \textit{et al} \([13]\).

Actually, Croft \textit{et al} \([13]\) did not use the (00L) series of Bragg peak to calibrate but only two reflections with \(K=1\) plus only two strong reflections (006) and (020) at high momentum transfer. That coarse calibration is presented in Fig. 10 of ref. \([13]\) in a log-log representation that we reproduced here in Fig. \(\bigstar\). We note first the saturation of the measured intensities for (020) strong Bragg reflection due to extinction effect. Another issue with this calibration is the sensitivity of the calculated structure factors to the oxygen content and the specific oxygen ordering of a given orthorhombic phase. This is particularly important for the K=1 Bragg peaks cross-section but also for the (006) reflection. A straightforward calculation shows that Bragg peak intensity in the (10L) or (01L) Bragg peaks cross-section varies considerably with the amount of actual oxygen atoms from \(x=6\) (tetragonal phase) to \(x=7\) (orthorhombic phase with full Cu-O chain) \([22]\). That is assuming that all extra oxygen atoms contribute to the cross-section to the long range lattice (as in ref. \([13]\)), but this ideal description does not correspond to reality due to imperfect oxygen ordering.

### TABLE I: Description for YBa\(_2\)Cu\(_3\)O\(_{6+x}\) samples studied using PND sorted by doping level. As discussed in \([3]\), the hole concentration \(p\) estimated from the \(T_c(p)\) relationships is given from ref. \([21]\) that Croft \textit{et al} \([13]\) are also using. [These estimates differ slightly from the ones originally given by Fauqué \textit{et al} \([1]\) where a simpler \(T_c(p)\) relation was used. These estimates from \([21]\) are more accurate but were not yet published at the time of Fauqué \textit{et al} \([1]\) was published]. The reported intensity of \(I_{\text{mag}}^{\text{orig}}\) is the magnetic intensity of the (011) reflection given in \([3]\). \(I_{\text{mag}}^{(011)}\) is the same quantity after correction from instrument resolution as discussed in section \(\bigstar\).

| Sample | ref. | doping p | \(T_c\) (K) | \(T_{\text{mag}}\) (K) | Oxygen ordering | detwinned | \(I_{\text{mag}}^{\text{orig}}\) (mbarn) | \(I_{\text{mag}}^{(011)}\) (mbarn) |
|--------|------|----------|-------------|----------------|----------------|-------------|----------------|----------------|
| A      | [1]  | 0.091    | 54          | 309 \(\pm\) 10 | n/a            | no          | 3              | 2.6            |
| H1     | [13] | 0.104    | 58          | -              | ortho-II       | yes         | -              | -              |
| B      | [1]  | 0.107    | 61          | 250 \(\pm\) 20 | ortho-II       | no          | 1.7            | 1.5            |
| M      | [2]  | 0.112    | 63          | 235 \(\pm\) 15 | ortho-II       | no          | 1.6            | 1.4            |
| C      | [1,5] | 0.115    | 64          | 220 \(\pm\) 20 | ortho-VIII     | yes         | 1.5            | 1.3            |
| H2     | [13] | 0.123    | 67          | -              | ortho-VIII     | yes         | -              | -              |
| D      | [1]  | 0.135    | 78          | 170 \(\pm\) 30 | n/a            | no          | 0.6            | 0.5            |
FIG. 1: (color online) Measured intensity of the nuclear Bragg peaks corrected by the Lorentz factor from sample H1 (ortho-II, p=0.104) of Croft et al.\textsuperscript{13} versus calculated structure factors ($|F_N|^2$). $|F_N|^2$ is calculated with two different models but using the same crystallographic data of Jorgensen et al.\textsuperscript{22}: (i) (open circles) all extra oxygen atoms contribute to the cross-section, $S(0) = 1$ in Eq. 2 as assumed by Croft et al.\textsuperscript{13}, (ii) (full circles) the finite correlation length along $c$ of the oxygen ordering is taken into account, corresponding to $S(0) \sim 1/3$ in Eq. 2. The dashed line is the same as the full line in Fig. 10 of ref. \textsuperscript{13}. Note that independently of these models, the structure factor for the (020) reflection has been underestimated by Croft et al.\textsuperscript{13} by a factor $\sim 2$.

There is an abundant literature about the different stages of oxygen ordering in YBCO (see e.g. \textsuperscript{18}). Typically, the oxygen ordering is characterized by a diffuse scattering with 2D ordering with large in-plane correlation length and short correlation range along $c\ast$. Only the ortho-II phase exhibits a three-dimensional ordering, the remaining ones are essentially two-dimensional\textsuperscript{13}. The extra oxygen form in-plane chains that do not become coherent along the $c$-direction, perpendicular to the CuO$_2$ plane. Actually, even in ortho-II phase, the extra oxygen atoms do not fully contribute to the lattice, explaining why the (011) cross section is measured larger than the simple calculation is giving.

More specifically, the nuclear structure factor for (01L) reflections can be written as,

$$\frac{d\sigma}{d\Omega}(01L) = |F_N|^2 = |b(L) - x b_O S(0)|^2$$  \hspace{1cm} (2)

where $b(L)$ is a sum of neutron coherent scattering length of the various atoms (Y,Ba,Cu,O) already present in the parent tetragonal compound, YBa$_2$Cu$_3$O$_6$. However, for these atoms, the structure factor is calculated using the atomic positions reported in the orthorhombic YBCO$_{6.5}$ phase\textsuperscript{22} to take into account the effect of the orthorhombic distortion. The contribution of the additional oxygen atoms in the Cu-O chains is not included in $b(L)$ but it is in the second term of Eq. 2 where $x = 0.54$ (for sample H1) is the extra oxygen content, $b_O$ is neutron coherent scattering length for oxygen, $S(0)$ is the prefactor of $S(Q)$, the scattering function of the extra oxygen atoms typically described by a Lorentzian function\textsuperscript{18}. By definition, $\int_{BZ} S(Q) d^Q = 1$ as the scattering function has to be normalized to 1 once integrated over the Brillouin zone (BZ). As shown in Eq. 2 there is a destructive interference between both terms. If $b(L)$ is large enough, the interference with the extra oxygen scattering is not very large and $|F_N|^2$ does not vary much with $x$. In contrast, if $b(L)$ is similar to $b_O$, the destructive interference cannot be neglected. However, if the order is 2D or at short-range, $S(0)$ is noticeably smaller than 1. For the larger correlation case considered in the ortho-II phase\textsuperscript{13}, $\xi_c = 58\,\text{Å}$, that corresponds to $S(0) \sim 1/3$. $b(L)=0.83$ barn is found the smaller for $L = 1$. Putting all these numbers together, one finds that $|F_N|^2 \simeq 0.54$ barn for the (011) reflection not 0.28 barn as quoted in Croft et al.\textsuperscript{13}.

In Fig. 1, the measured intensities are reported versus the calculated structure factor, $|F_N|^2$, for these two limits : (i) the long range oxygen ordered ortho-II phase considered by Croft et al, $S(0) = 1$ and (ii) the short range oxygen ordered ortho-II phase with $\xi_c = 58\,\text{Å}$, $S(0) \simeq 1/3$. The figure shows that the expected linear behavior (dashed line) works as good for both models (only with three points as the (020) reflection is affected by extinction effect). Therefore, Fig. 1 does not validate the structural model with $S(0) = 1$, in contrast to the suggestion of Croft et al\textsuperscript{13}. The H1 ortho-II sample has been studied with hard x-ray diffraction measurements\textsuperscript{14} where the oxygen-chain order is reported to have a finite coherence length along c, $\xi_c = 55\,\text{Å}$ corresponding to $S(0) \simeq 1/3$. In ortho-VIII phase, the oxygen ordering is essentially 2D, so, $S(0)$ is even smaller and $|F_N|^2$ larger. That underestimation of $|F_N|^2$ of Croft et al\textsuperscript{13} yields an additional overestimation of the expected magnetic scattering by a factor 2.

In this section we show an overestimation of the IUC intensity in Croft et al\textsuperscript{13} by factor 2 from the difference in doping (section II A). Another factor of more than 2 arises from twin-averaging $vs$ fully detwinned sample (section II B) that applies only for the (010) Bragg spot. Finally, a last factor of 2 is due to data calibration (section II C) which occurs for the (011) reflection where the value of $b(L)$ is smaller. Therefore, the overall conclusion of this section is a systematic overestimation by a factor of $\sim 4$-5 of the expected magnetic scattering (quoted "Fauqué et al") in the figures 8,9 and 11, of Croft et al\textsuperscript{13}, making the conclusion of lack of observation of our observed signal invalid.
Next we examine the different data collection strategies. As the expected magnetic signal is weak compared to the leakage of the nuclear intensity due to imperfect polarization, $p \sim 0.95-0.96$, of the instruments, the only way to observe the signal is to perform a very accurate temperature dependence of the flipping ratio ($R$), i.e. $R = \frac{I_{NSF}}{I_{SF}}$ of a given Bragg spot ($I_{NSF}$ and $I_{SF}$ stand for the non-spin-flip and spin-flip intensities, respectively). The expected signal can show up above a reference line (called baseline) which has to be determined at high temperature (above $T_{mag}$). Unfortunately, only 1 or 2 points have been measured by Croft et al [13] above the reported ordering temperature $T_{mag}$ (which coincides with the pseudogap temperature, $T^*$). This is unfortunately insufficient to properly characterize the shape of that baseline and distinguish it from a genuine magnetic signal.

Further, Croft et al [13] did not perform the same method to collect the data as the one used in previous PND studies. They used a more text-book way which consists of realigning the sample at each temperature, implying that the spectrometer has to move at every temperature. This method would be very reliable if the outcome of the measurements were not affected by erratic positioning errors of the spectrometer at each alignment. We actually discarded that method rapidly as it did not give reliable enough flipping ratios values compared to the required accuracy. This is due to the limit of mechanical stability of the instruments. It adds additional systematic and random errors in the countings. Although Croft et al [13] do not report a very large distribution of countings between each given temperature, this can be nevertheless seen in their data where some of their points exhibit clear departure from the expected statistical errors of the counting. As the total number of points as a function of the temperature (6-8) for each curve is rather scarce, this is detrimental to get confident determination of both the shape of the baseline and possible magnetic signal on top of it, especially when the point at 300 K is the point which drifts away from the expected statistics.

Croft et al [12] argued that it is crucial to realign the sample at every temperature as the lattice parameters change with temperature and that the flipping ratio is sensitive to A4 ($\sim 2\theta'$-scan) (Fig. 6d in [13]). We are aware of this possible issue but we show that this effect is not crucial. This argument put forward by Croft et al leads to some inconsistency. Assuming that the argument were correct, then one should detect an artefact magnetic signal on any Bragg reflection. This is not what we have observed [15]. Furthermore, this signal should be stronger at large momentum transfer, such as for the (02L) reflections where no effect was reported [2, 3]. Indeed, the effect should be about twice larger at large $Q$ such as the (020) reflection than at lower $Q$ for the (01L) reflections. Precisely, the effect is 2.4 less for (010) compared to (020) as it is in the ratio of $K/\cos(\theta)$ of both Bragg peaks indexes. This is not observed in any of our data (see e.g. [2, 3]). Furthermore, under this assumption, there is no way to understand how an artificial magnetic signal could fulfilled the magnetic sum rule obtained by the neutron polarization analysis [1, 2]. Croft et al [12] are incorrect when they wrote in their section III.C “(similar changes are expected for the (010) and (011) reflections)”. It should be stressed that this effect is less important in configurations of broad momentum instrument resolution, for instance in the case of large samples with large mosaicities which broadens the instrument resolution.

In order to keep a highly stable flipping ratio, previous PND studies rule out the full realignment of the sample at each temperature. This is why we have way better accurate flipping ratio, a keypoint of success to determine a genuine magnetic signal. However, we realign the rocking ”A3” angle at each temperature if necessary. This has less impact of the flipping ratio stability. Our strategy to collect the data then consists of measuring a lot of points in temperature with high statistics with no spectrometer movement. It should be stressed that Croft et al [13] do not report any data obtained this way although we have established that this is a proper technique to disentangle unwanted shift of the flipping ratio with temperature (the change of the ”baseline”) from a genuine magnetic signal [11].

Further, they should have considered the possibility of a drift of the baseline due to the fact that the sample move up and down as a function of temperature, following the construction and dilation of the cryostat sample stick on which the sample is attached. Therefore, the sample is displaced vertically in temperature and thus probes different flipping ratios due to inevitable polarization inhomogeneities of the beam because of large monochromator and analyzer sizes [11]. This would be especially the case for a small sample compared to sample stick shrinking as Croft et al [13]. Therefore, their claim that ”the neutrons always emanate from the same part of the monochromator and strike the same part of the analyzer,” cannot be correct when changing temperature.

Incidentally, another experimental issue in [12] is related to the incoherent background subtraction, which should be measured away from the Bragg position. Again, a different strategy has been applied compared to the previous PND studies in [3, 5, 4, 11]. We prove that this background subtraction to be important for a quantitative analysis of the data when the magnetic signal is weak [11]. No background subtraction was made in the original reports [1, 2]. The choice in momentum space of the reference background wavevector is important as it has to be a location where no magnetic signal is present. We choose points by shifting the in-plane momentum transfer such as $Q = (0,0,9,1)$ for the reflection (011). Typically, on a triple axis instrument, the SF incoherent background intensity at $Q = (0,0,9,1)$ slightly increases upon cooling (negative slope with temperature), following the expected behaviour of a Debye-Waller factor. Using po-
larization analysis, we prove that magnetism at such a location is weaker by two order of magnitude (see e.g. [4]).

Instead, Croft et al [13] choose $Q = (0.1, 0.9)$ along $c^*$ for the background subtraction and did not mention any polarization analysis to address possible magnetism at this $Q$-location. Near optimal doping, we show that magnetic signal is not negligible at this $Q$-location [4] due to short-ranged IUC correlations along $c$. Therefore, that choice is particularly a bad idea as it would necessarily reduce possible intrinsic IUC signal. Actually, Croft et al [13] report the intensity at $Q = (0.1, 0.9)$ in Figs. 7b-c in a logarithm scale. Interestingly, it occurs that the SF intensity decreases upon cooling (positive slope with temperature) at this $Q$-location (this is especially clear for the H1 sample, Fig. 7b). This implies one of two possibilities: either, this sloping background is the true incoherent background (and then it should be removed from the Bragg peak (011) SF intensity), or it suggests a vanishing at low temperature of a magnetic signal present at high temperature at this $Q$-location. This latter possibility is related to the fact that the positive slope with temperature background behaviour contrasts with the known behaviour of the incoherent SF background (negative slope with temperature) discussed above. Both possibilities leads to interesting opportunities. Croft et al [13] made no attempt to analyse this data along these lines, only fitting it by a constant.

Finally, no attempt of polarization analysis is reported in [13] to search for the weak magnetic signal at the Bragg reflection (011) or to accurately determine an upper limit for a magnetic scattering intensity. It should be strongly emphasized that previous PND studies reported a magnetic signal at very specific Bragg reflections (the signal was absent on others) and complementary polarization analysis demonstrate unambiguously the magnetic nature of the observed scattered intensity [1, 2, 6, 7, 10].

As a result of the various points discussed above, Croft et al [13] cannot estimate the shape of the "baseline" as they can only rely on the point measured at 300K to re-scale their data (even if sometimes that point is not included in their fitting line). That necessarily alters the data accuracy and degrades the threshold of detection of a small signal.

IV. EFFECTS OF SHORT-RANGE MAGNETIC CORRELATIONS

It has been already discussed by Fauqué et al [11] that local probe measurements had not observed a magnetic signal consistent with our measurements. Typically, zero-field muon spin resonance ($\mu$SR) [23, 24] and Nuclear Magnetic Resonance (NMR) [23] experiments in YBCO found no evidence for a static and long range magnetic order. First, let us recall that the situation with $\mu$SR experiments has been contradictory (see [11] for an extensive discussion). Second, it was envisaged [1, 24] that the magnetic moments could fluctuate slowly enough to appear static to neutrons, but too fast enough to be identified as a magnetic order in $\mu$SR and therefore even more for NMR which probes even slower timescales. Recently this picture has been confirmed as slow magnetic fluctuations have been discovered in YBCO samples [14] using a longitudinal field $\mu$SR technique. These magnetic fluctuations are related to our magnetic order as a critical slowing down [14] is also reported at the same temperature $T_{mag}$ where the neutron magnetic signal sets in. Beyond the YBCO bilayer system, new set of $\mu$SR data in the bilayer system, $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$+$\delta$, is also found to support the existence of the magnetic signal [15].

Croft et al [13] dismiss these recent developments and efforts made to refine the $\mu$SR studies. The conclusion of the PND experiment of Croft et al is no more supported by the most recent $\mu$SR studies. J. Zhang et al [14] found that magnetic correlations fluctuate, $\sim 10$ ns timescale, even for strongly underdoped samples as the ones discussed here. These low frequency fluctuations are necessarily related to the formation of finite-size magnetic domains [25], corresponding to finite correlation lengths of the magnetic order observed in neutron diffraction. Such a short range correlation length of the IUC order has been observed [3] near optimally doped YBCO sample. Note that when the correlation length becomes too short, such as in $\text{La}_{2−x}\text{Sr}_x\text{CuO}_4$ [8], the magnetic signal cannot be observed anymore on the Bragg reflection.

In underdoped samples, for a doping $p\approx 0.11$ (samples A and M in Tab. 1), we observed resolution limited magnetic peaks [1, 2]. However, the PND experiment was performed on large single crystals with large mosaicity, corresponding to rocking scans with a full width at half maximum (FWHM) of $\sim 1^\circ$, implying that correlation length $\xi \geq 75$ Å. The rocking "A3" scan is similar than a scan along L. On Bragg reflections, (010) or (020), a rocking scan matches a transverse scan, i.e. a scan along the (001) direction. Instead, the measurements carried out by Croft et al [13] are performed on small samples with high mosaic spread yielding an observed rocking scan FWHM of $0.35^\circ$ (Fig 6 and 7). Therefore, if the magnetic order exhibited a finite correlation length $\xi = 75$ Å, the magnetic signal would be characterized by a FWHM of $\sim 0.95^\circ$ in the A3-scan, with a peak intensity reduced by a factor $\sim 3$ in comparison with a resolution limited signal, like a nuclear Bragg peak. A sketch of these two situations is reported in the top-right panel of Fig. 4. The magnetic amplitude at the Bragg position would be accordingly $\sim 3$ times reduced compared to the one of true long range ordered state.

In conclusion of this section, in case of short range magnetic correlations, for example $\xi = 75$ Å, the magnetic signal would be characterized by a FWHM of $\sim 0.95^\circ$ in the A3-scan, with a peak intensity reduced by a factor $\sim 3$ in comparison with a resolution limited signal, like a nuclear Bragg peak. A sketch of these two situations is reported in the top-right panel of Fig. 4. The magnetic amplitude at the Bragg position would be accordingly $\sim 3$ times reduced compared to the one of true long range ordered state.
FIG. 2: (color online) Measured intensity of the nuclear Bragg peaks corrected by the resolution factor $R_0(Q)$ versus the calculated structure factors ($|F_N|^2$) for the sample C detwinned [1]. The data has been measured on the triple-axis 4F1 at Orphée reactor (CEA Saclay). The structure factors are calculated using crystallographic data in [22]. The dashed black line gives the calibration of the data in barn (Eq. 3). The dashed red line corresponds to the calibration used in [1] (7 barns for the (004) reflection). That yields a renormalization by $\sim 15\%$ of the intensity in absolute units compared to our previous report [1].

V. IMPROVED DATA ANALYSIS

Finally, motivated by the fact that a detailed comparison is necessary to appreciate the possible differences between the study of Croft et al [13] with earlier PND works, we refine the data calibration procedure of Fauqué et al [1]. Originally, the data were calibrated onto the Bragg peak reflection (004) but the instrumental resolution was neglected as a subsidiary effect. We then reassess below the complete data calibration of the results of the detwinned high-quality sample C [1].

To perform the calibration, one needs to estimate the instrumental resolution correction. The resolution of a triple-axis spectrometer is known to be described by a 4D (3 directions for momentum transfer and one for energy) Gaussian function [27]. The scattering intensity is the convolution of the neutron scattering cross section with the instrument resolution ellipsoid. A Bragg peak is a Dirac function in phase space of both the momentum transfer and the energy. For a Bragg scattering, the measured intensity at $Q = (HKL)$ then reduces to the product of the computed structure factor with the volume of the resolution ellipsoid. That reads:

$$I(Q) = R_0(Q)|F_N|^2$$

(3)

$R_0(Q)$ is determined by the normalization of the spectrometer 4D Gaussian resolution [28]. In general, for a triple-axis spectrometer [29], $R_0(Q)$ is never as simple as the Lorentz factor, $1/\sin(2\theta)$, used for a diffractometer (two-axis instrument, no analysis of the scattered neutron energy). Croft et al [13] are then not correct to apply the Lorentz factor in their calibration curve (fig. 10) even for A3-integrated intensities. To determine $R_0(Q)$, we use a Cooper-Nathans [27] formalism but with the use of mosaic spreads of the diffracting crystals and of the divergence angles given by the beam geometry (distances between the different elements composing the instrument, sizes of crystals,...).

For the detwinned sample C [1], one can compute the proper resolution factor $R_0(Q)$. Fig. 4 shows the comparison of the measured intensity of the nuclear Bragg peaks divided by the resolution factor $R_0(Q)$. The figure shows canonical behaviour of a calibration curve (see an example, Fig. 1 in [31], in the context of cuprates): i) the expected linear behavior given by Eq. 3 at small structure factor and ii) a saturation at larger structure factor due to extinction effects. From this linear calibration at small enough $|F_N|^2$ (black dashed line), one obtains a renormalization of about 15% for the magnetic cross-section for sample C of 1.3 ± 0.1 mbarn at (011) reflection compared to our previous estimate (red dashed line). As the IUC magnetic intensity at the reflection (011) does not depend much on the sample detwinning [2], one can report the corrected intensity for different samples versus the doping level (Fig. 5) from the published data [1,3]. As the intensity is proportional to the square of

FIG. 3: (color online) Doping dependence of the IUC ordering temperature $T_{mag}$ and magnetic cross-section of the Bragg reflection (011). The neutron cross-section for the IUC order is estimated at a temperature $\sim 10K$ above $T_c$. Data are taken from refs. [1–5].
FIG. 4: (color online) $1/R = I_{SF}/I_{NSF}$ data for sample H1 (p=0.104) reproduced from Croft et al [13]. The comparison in dashed lines is made with the results of sample B of [1] (see Tab. I). The sample B is a twinned sample but the data have been corrected to correspond to the case of a twin free sample. Three different scenarios are considered (see text). The top right panel represents A3 rocking scans convoluted by the resolution function (blue) for the scenario B (red) for the scenario C.

The inverse of flipping ratio at the reflection (020) is linear in temperature with a positive slope as it has been shown to exist [2, 5]. As discussed in section III, that slope is inevitable as the sample is displaced in the neutron beam upon cooling. With the large grey shaded area, we represent the zone of the limit of detection. This area is due to combined effects of the statistical errors of each points, occurrence of off-statistical points (possibly related to errors in positioning as discussed in section III) and the scarce number of points. This area is simply deduced from the measurements at the Bragg (020) reflection where no magnetic signal is expected for the IUC order. For clarity, the same error of $\delta R^{-1} \approx \pm 0.001$ is defined for both figures. That error is typically equivalent to an error of $\delta R \approx \pm 2$ on the flipping ratio or $\delta I_{SF} \approx \pm 4\%$ of the spin-flip intensity. This uncertainty is not negligible as the highest ratio of the IUC intensity to the spin-flip total intensity, that we have reported, is only $\sim 10\%$ for detwinned sample [1, 2, 11] and about $\sim 3-4\%$ otherwise [1, 2] for the corresponding doping levels. The error on the spin-flip intensity in previous PND...
experiments [1, 2, 3] ranges between $\delta I_{SF} \simeq \pm 0.5 - 1 \%$. (see e.g. Figure S1 in Supplemental materials of Mangin-Thro et al [3] for an estimate of $\delta R^{-1}$).

On Figs. 4 and 5, the detection limit of $\delta R^{-1} \simeq \pm 0.001$ is next reported for the Bragg reflections (010) and (011) where the magnetic signal is expected. To compare that set of data, with those of Fauqué et al. [1], we then consider three different scenarios which are plotted in Figs. 4-5: A) no magnetic signal is present, B) a long range magnetic order is present with the amplitude reported in [1] C) a magnetic order with short range correlation along the c-direction is present with the amplitude reported in [1]. For the sample H1, one compares with the amplitude measured for sample B (see table 1) assuming that this sample would be detwinned for a proper comparison. For the sample H2, one compares with the amplitude measured for sample C. The data have been normalized in absolute units using the structural model discussed in section II C of short range oxygen ordering.

One positive conclusion that could be derived from the study of Croft et al. [13] is that the use of sample with a very good mosaic has raised question concerning the length of the magnetic correlations for the IUC order. Short ranged correlations were already established in YBCO around optimal doping [1] but not for more underdoped samples. Put together with the recent $\mu$SR studies [14, 15], this could reinforce the idea according to which there could be finite size magnetic domain, with very slow dynamics. The role of the finite size correlations and related forward scattering was recently addressed by C.M. Varma [26] within the loop current framework.

Finally, it is worth pointing out that independent theoretical ideas have been developed to interpret our measurements. Firstly, the magnetic order is generally described by microscopic loop currents within each unit cell [16, 31] and is therefore usually associated with orbital magnetic moments. Further developments of the loop current theory have been made to account for additional experimental facts [17, 26]. Recently, other types of loop currents have been discussed as well [32] where translationally-invariant states with topological order coexisting with both Ising-nematic order and spontaneous charge currents. Following a different path, magnetic quadrupoles on the Cu ions have been also proposed to account for our results [33] with possible microscopic mechanism [34]. Further, we recently reported similar time-reversal broken symmetry in iridates [35] implying the possible generalization of loop current-type electronic phase in other oxides. In both iridates [36] and cuprates [37], second harmonic generation optical measurements found an odd-parity magnetic order parameter exactly in the same temperature and doping ranges, fully consistent with the loop current-type phase. In cuprates, other experimental techniques confirm the same broken symmetry in the pseudogap phase, either time reversal symmetry broken symmetry [38], or loss of both C4 rotation and mirror symmetry in optical birefringence in YBCO [39].

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