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## X-Ray Diffraction Observations of a Charge-Density-Wave Order in Superconducting Ortho-II $\text{YBa}_2\text{Cu}_3\text{O}_{6.54}$ Single Crystals in Zero Magnetic Field

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X-ray diffraction measurements show that the high-temperature superconductor  $\text{YBa}_2\text{Cu}_3\text{O}_{6.54}$ , with ortho-II oxygen order, has charge-density-wave order in the absence of an applied magnetic field. The dominant wave vector of the charge density wave is  $\mathbf{q}_{\text{CDW}} = (0, 0.328(2), 0.5)$ , with the in-plane component parallel to the  $\mathbf{b}$  axis (chain direction). It has a similar incommensurability to that observed in ortho-VIII and ortho-III samples, which have different dopings and oxygen orderings. Our results for ortho-II contrast with recent high-field NMR measurements, which suggest a commensurate wave vector along the  $\mathbf{a}$  axis. We discuss the relationship between spin and charge correlations in  $\text{YBa}_2\text{Cu}_3\text{O}_y$  and recent high-field quantum oscillation, NMR, and ultrasound experiments.

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Charge density waves (CDWs) have recently been observed in the high-temperature superconductors (HTSs)  $\text{YBa}_2\text{Cu}_3\text{O}_y$  (YBCO) and  $(\text{Y}, \text{Nd})\text{Ba}_2\text{Cu}_3\text{O}_y$  [1–3]. The CDW, which competes with HTSs, develops in a region inside the celebrated pseudogap phase, where a number of other probes, including high-field NMR [4], the Kerr effect [5], and the Hall effect [6], show signatures of electronic ordering. This last effect is smoothly connected to the low-temperature high-field quantum oscillations (QOs) [7] that demonstrated the existence of small Fermi surface pockets.

The existence of ground states with competing order is central to many theories of HTSs. A widely discussed example is “stripe order,” that is, a state with coexisting charge and spin order [8]. Stripe order is observed in some HTSs and related compounds, such as  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$  [9] and  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  [10]. It is important to establish whether the tendency toward stripes is a generic property of the cuprates and whether the spin and charge correlations are always related.

$\text{YBa}_2\text{Cu}_3\text{O}_y$  differs from, e.g.,  $\text{La}_{2-x}(\text{Ba}, \text{Sr})_x\text{CuO}_4$ , in that it contains bilayers of  $\text{CuO}_2$  planes, separated by layers containing a certain fraction (depending on  $y$ ) of Cu-O chains. The oxygen-filled chains, which run along the orthorhombic crystal  $\mathbf{b}$  direction, tend to order and are labeled ortho- $N$ , depending on the repeat length ( $N$ ) of the ordering of the chains along  $\mathbf{a}$  [11–13]. A major gap in the CDW picture to date was the failure to observe a CDW in the ortho-II state (the most highly ordered state,

having alternately full and empty Cu-O chains). This was surprising because many studies of this composition had suggested that such an order is present, at least in a high field [4,6,14–17]. Most recently, ultrasound measurements have indicated a two- $\mathbf{q}$  state in high magnetic fields [18].

In this Letter, we report the observation of a CDW in an ortho-II sample of  $\text{YBa}_2\text{Cu}_3\text{O}_{6.54}$  in a zero magnetic field with dominant wave vector  $\mathbf{q}_{\text{CDW}} = (0, 0.328(2), 0.5)$ . This contrasts with both lower and higher dopings, where two wave vectors are observed corresponding to two modulations of similar amplitude [1–3]. Both the wave vector magnitude and the direction of  $\mathbf{q}_{\text{CDW}}$  differ from those inferred from NMR measurements in a high field [4]. The propagation vectors of the charge and spin correlations in ortho-II YBCO do not appear to follow the simple relationship  $\delta_{\text{charge}} = 2\delta_{\text{spin}}$  observed in stripe systems [9,10]. We examine the doping dependence of the CDW order in high quality ortho-II, III, and VIII samples (hereafter denoted as o-II, o-III, and o-VIII).

We carried out high energy (100 keV) x-ray diffraction experiments on three  $\sim 99\%$  detwinned  $\text{YBa}_2\text{Cu}_3\text{O}_y$  single crystals (sample characteristics are given in Table I). These samples have orthorhombic crystal structures [ $a \approx 3.82$  (ignoring the chain-ordering superlattices),  $b \approx 3.87$ , and  $c \approx 11.7$  Å]. The o-II sample shows a more perfect oxygen-chain order than the other compositions. The coherence lengths are  $\xi_a \sim 110$  Å,  $\xi_b > 150$  Å and  $\xi_c = 55$  Å for o-II,  $\xi_a \sim 20$  Å and  $\xi_c < 10$  Å for o-III, and

TABLE I. Characteristics of the  $\text{YBa}_2\text{Cu}_3\text{O}_y$  samples studied. The superconducting  $T_c$  was determined from the 1 Oe field-cooled magnetization, and doping was evaluated from Ref. [19].  $T_{\text{CDW}}$ ,  $\delta_1$ , and  $\delta_2$  are derived from our high energy x-ray experiments—see also the note added at the end of this Letter—(quoted uncertainties in the values of  $\delta$  are dominated by minor crystal alignment errors) and from Ref. [3].

y in YBCO	Oxygen ordering	Doping level $p$	$T_c$ (K)	$T_{\text{CDW}}$ (K)	$\delta_1(\mathbf{a})$ (r.l.u.)	$\delta_2(\mathbf{b})$ (r.l.u.)
6.54	o-II	0.104	58	155(10)	0.320(2)	0.328(2)
6.67	o-VIII	0.123	67	140(10)	0.305(2)	0.314(2)
6.75	o-III	0.132	74	140(10)	0.30 [3]	0.308(2)

$\xi_a \sim 25 \text{ \AA}$  and  $\xi_c < 10 \text{ \AA}$  for o-VIII. Our samples are highly uniform, strongly indicating that the CDW occupies the entire sample volume and not a fraction.

Samples were mounted in a closed-cycle cryostat on a four-circle diffractometer on beam line BW5 at the DORIS storage ring (DESY). This allowed access to a wide range of reciprocal space ( $h, k, \ell$ ) expressed in units of ( $2\pi/a, 2\pi/b, 2\pi/c$ ) at temperatures down to 6 K.

A CDW modulation with characteristic wave vector  $\mathbf{q}_{\text{CDW}}$  gives rise to satellites around reciprocal lattice points at positions  $\mathbf{Q} = \boldsymbol{\tau} \pm \mathbf{q}_{\text{CDW}}$ . High energy x-ray diffraction is sensitive to the atomic displacements parallel to the scattering vector  $\mathbf{Q}$  [2]. Our previous measurements [2] on o-VIII showed  $\mathbf{q}_{\text{CDW}} = (\delta_1, 0, 0.5)$  and  $(0, \delta_2, 0.5)$  (see Table I). Figures 1(a) and 1(b) show  $k$  scans performed on YBCO o-II through the positions  $(0, \delta, \ell)$  and  $(0, 2 - \delta, \ell)$ , with  $\ell = 0.5$  and  $6.5$  and  $\delta \sim 0.3$ . No CDW peaks were found at  $\ell = 0.5$ , but well-defined peaks were observed at  $\ell = 6.5$ . These peaks constitute the first direct x-ray evidence for CDW order in YBCO o-II and indicate that the displacements are mainly polarised along  $\mathbf{c}$ . For comparison,  $k$  scans through the positions  $(0, 2 - \delta, \ell)$  in o-VIII

and o-III are shown in Figs. 1(c) and 1(d) for  $\ell = 0.5$  and  $6.5$ ; again, for modulations along  $k$ , the signal at  $\ell = 6.5$  is stronger.

In YBCO o-VIII and III, a lattice modulation is found along both the  $\mathbf{a}$ - and  $\mathbf{b}$ -axis directions [1–3]. For o-II, we have searched for a CDW along the  $\mathbf{a}$  axis, concentrating on positions where a signal was observed in o-VIII. Wave vectors  $(|n - \delta|, 0, \ell)$  with  $n = 2, 4$  for  $\ell = 0.5$  and  $n = 0, 2, 4$  for  $\ell = 6.5$  were measured, and no signal from a lattice modulation was found above the noise level [see Figs. 2(a) and 2(b)]. However, see also the *Note added in proof* at the end.

Even though the CDW structures in YBCO o-II, III, and VIII are different, the signals from the  $\mathbf{b}$ -axis modulation have a very similar dependence on  $\ell, T$ , and the magnetic field. Figure 3(a) shows the  $\ell$  dependence near  $\ell = 6.5$  at  $T \sim T_c$ . All three compounds have a broad peak at  $\ell \approx 6.5$ , with a width corresponding to a correlation length of  $\xi_c \leq 10 \text{ \AA}$ . The  $T$  dependence of several reflections is plotted in Fig. 3(c). As was previously shown for YBCO o-VIII and o-III [1–3], the intensity grows below an onset temperature  $T_{\text{CDW}}$  ( $\sim 140 \text{ K}$ ) down to the respective  $T_c$ 's, below which a partial suppression takes place. Here, we show a similar behavior in o-II but with a slightly higher onset temperature. The application of a magnetic field enhances the low- $T$  intensity in a similar fashion to that reported in YBCO o-VIII [2]. It is interesting to note [see Fig. 3(b)] that the width of the Bragg peak becomes smaller in a field with 11.5 T along  $\mathbf{c}$ , corresponding to an increase of correlation length  $\xi_b$  from  $48 \pm 9$  to  $63 \pm 7 \text{ \AA}$ . The presence of a magnetic field suppresses superconductivity, and the resurgent competing CDW increases its correlation length.

Modeling of our  $\mathbf{b}$ -direction data for all three dopings suggests that the displacement pattern involves  $\mathbf{c}$ -axis

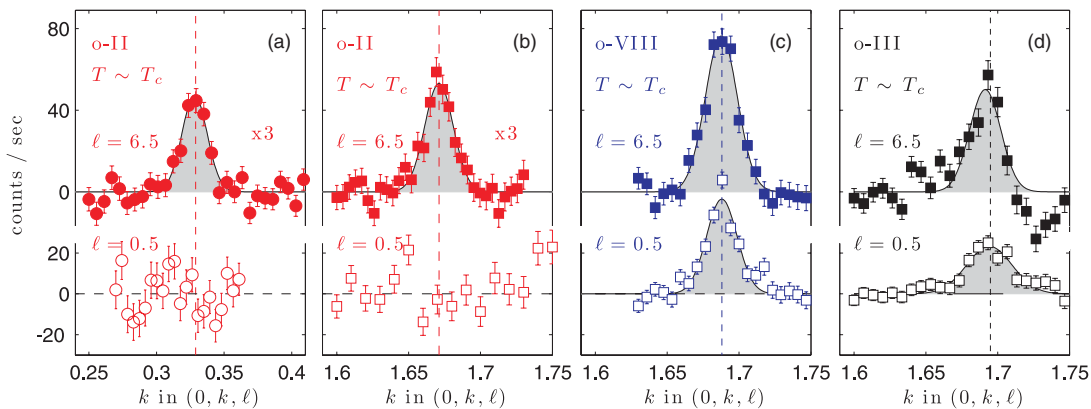


FIG. 1 (color online).  $k$  scans through the CDW wave vector positions  $(0, \delta_2, \ell)$  and  $(0, 2 - \delta_2, \ell)$  in YBCO o-II, VIII, and III at  $T \sim T_c$ . (a),(b) For o-II,  $\delta_2 = 0.328(2)$  incommensurate peaks are only observed at  $\ell = 6.5$ , suggesting a strong CDW displacement along  $z$ . (c),(d) For o-VIII and o-III, peaks are observed at  $\ell = 0.5$  and  $6.5$  with  $\delta_2 = 0.314(2)$  and  $0.308(2)$ , respectively. Linear backgrounds have been subtracted in (a) and (b). To avoid contamination from weakly  $T$ -dependent spurious peaks in (c) and the o-III superlattice peak from the minority domain in (d), we plot  $I(T_c) - I(140 \text{ K})$ , corrected for a sloping background, as in (a) and (b). Intensities in (a) and (b) have been multiplied by a factor of 3 to compensate for the smaller o-II sample size.

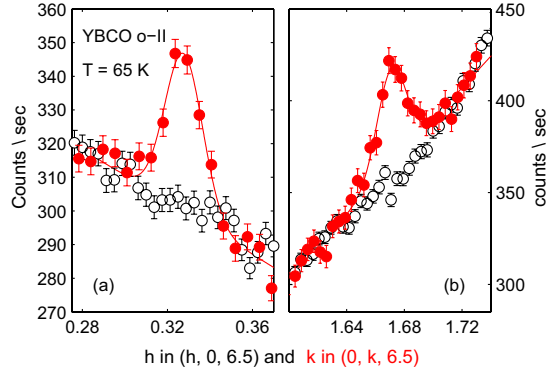


FIG. 2 (color online). (a),(b)  $h$  and  $k$  scans, taken on o-II, through  $(|n - \delta_1|, 0, 6.5)$  and  $(0, |n - \delta_2|, 6.5)$  with  $n = 0, 2$  and  $T \sim T_c$ . The  $k$  scans (filled red circles), showing lattice modulation peaks at  $(0, \delta_2, 6.5)$  and  $(0, 2 - \delta_2, 6.5)$ , are the same as those displayed in Figs. 1(a) and 1(b). Equivalent measurements in the  $(h, 0, \ell)$  plane (open black circles) reveal no evidence for a lattice modulation at  $\delta_1 \sim \delta_2$ . Notice that the lattice modulation peaks are 2 orders of magnitude weaker than the reflections from the ortho-II structure.

displacements of the bilayer oxygens, similar to those proposed for the soft phonon in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  [20]. There are small differences between the values of the modulation periods for the **a** and **b** directions for all three compounds (Table I) and in the patterns of atomic displacements, showing that the influence of the chains on the planes is also noticeable in o-VIII and o-III. If the two distortions

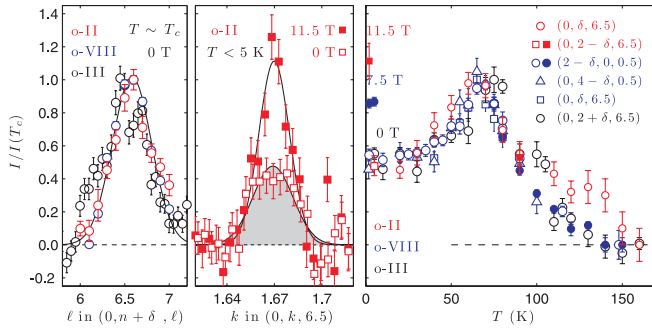


FIG. 3 (color online). Out-of-plane momentum  $\ell$ , field, and temperature dependences of the CDW modulation peaks found in YBCO o-II (red symbols), o-VIII (blue symbols), and o-III (black symbols). All intensities have been background subtracted and normalized to  $I(T_c)$  in a zero field. (a)  $\ell$  dependence of the peak height of  $k$  scans through  $(0, n + \delta_2, \ell)$ , with  $n = 0$  for o-II and o-VIII and with  $n = 2$  for o-III. All compounds show a broad peak centered at  $\ell \sim 6.5$  and a  $c$ -axis correlation length  $\xi_c$  comparable to that previously reported [2] in o-VIII at  $T = 2$  K for  $\ell = 0.5$ . (b) Measurement in a separate cryostat of the effect on CDW intensity in o-II of a magnetic field applied with a component 11.5 T along the  $c$  axis of the crystal. (c) Temperature dependence of peak intensities, measured at the wave vectors indicated. The filled symbols are data taken in a magnetic field.

develop independently, one would expect a different  $T_{\text{CDW}}$  for modulations along each direction, with the postulated two- $\mathbf{q}$  state forming at lower temperatures. To date, we have no evidence for this, although observations in o-II most clearly indicate a difference between the CDW order in the **a** and **b** directions. The in-plane electronic anisotropy in YBCO arises from the chains; thus, they must ultimately be responsible for this difference between the **a** and **b** directions. An obvious mechanism is through the chain Fermi surface [21] with spanning vectors along  $\mathbf{b}^*$ , which might encourage CDW formation. We note that STM observations on the chain surface of optimally doped YBCO [22] show such behavior, with a  $\delta_2 \sim 0.3$ . Furthermore, the alternating filled and empty chains create an additional potential which would fold the Fermi surface along  $\mathbf{a}^*$  and thereby change the band structure [21].

An important issue in the cuprates is the relationship of the spin and charge correlations [1,2,9,10], where the underlying antiferromagnetism (AFM) and charge density have modulations characterised by wave vectors  $\delta_{\text{spin}}$  and  $\delta_{\text{charge}}$ , respectively. In a simple stripe picture of intertwined spin and charge correlations [10], these yield spin and charge peaks at positions  $\tau_{\text{AFM}} \pm \delta_{\text{spin}}$  and  $\tau_{\text{lattice}} \pm \delta_{\text{charge}}$ , where  $\delta_{\text{charge}} = 2\delta_{\text{spin}}$ . This simple relationship appears to describe observations in  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$  (see Fig. 4) and  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  [9,10]. In YBCO, the low-frequency spin fluctuations are anisotropic [23,24], with the strongest response for  $\delta$  along  $\mathbf{a}^*$ . Indeed, lightly doped  $\text{YBa}_2\text{Cu}_3\text{O}_y$  shows magnetic order [25] with  $\delta$  along  $\mathbf{a}^*$ . Thus, in YBCO (see Fig. 4), not only are  $\delta_{\text{spin}}$  and  $\delta_{\text{charge}}$  in different directions, but they show different trends, and  $|\delta_{\text{charge}}| \neq 2|\delta_{\text{spin}}|$ . These differences suggest

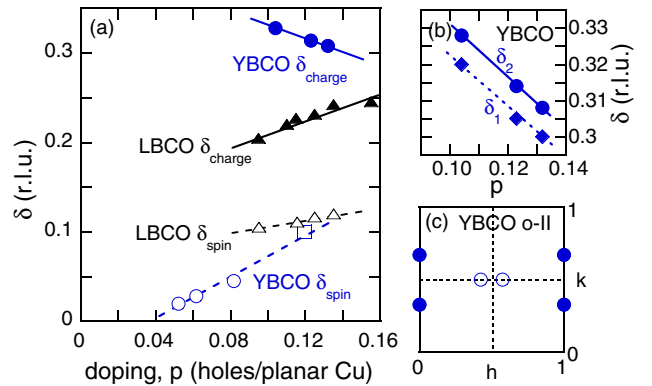


FIG. 4 (color online). (a) Spin and charge incommensurability versus doping for YBCO and  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$  (LBCO). The spin incommensurability of both YBCO [25] and  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$  [9] increases with doping. In LBCO, the spin and charge incommensurability are simply related:  $\delta_c \approx 2\delta_s$ . In YBCO, the spin and charge incommensurability have opposite trends with doping. (b) The charge incommensurability in YBCO, plotted on an expanded scale. (c) In YBCO o-II, the dominant wave vectors of the spin ( $\delta_{\text{spin}}$ ) and charge ( $\delta_{\text{charge}}$ ) modulations are along different directions: the **a** and **b** axes, respectively.



that  $\delta_{\text{spin}}$  and  $\delta_{\text{charge}}$  have different origins; e.g., they may be determined by different Fermi surface nesting vectors.

We may also consider  $T_{\text{CDW}}$  as a function of doping (Table I), although we stress the difficulty in determining unambiguously the onset of a small signal from a large background. In Fig. 3(c), it is clear that, for o-II, the CDW appears at a higher temperature, indicating that, as the hole doping is reduced,  $T_{\text{CDW}}$  increases. This pattern coincides roughly with the onset of the polar Kerr effect [5] and the inflection point  $T_H$  below which the Hall coefficient begins to fall toward negative values [6], as has been suggested previously [2]. This indicates that all these measurements are sensitive to the same lattice symmetry breaking process.

This places our observations in context with other low-field measurements. However, originally, the existence of CDW order was indicated by high-field NMR work on o-II [4]. This was revealed by a splitting, at a high magnetic field and low temperature, of the NMR lines of the Cu2F sites, which lie in the CuO<sub>2</sub> planes next to a filled CuO chain. This splitting was most simply explained by invoking a one- $\mathbf{q}$  CDW along the  $\mathbf{a}$  direction with  $\delta = 0.25$  [26], which could give a Cu2F splitting while having no effect on the Cu2E sites next to empty chains. However, very recent ultrasonic data [18] indicate that, under similar high-field conditions, o-II is actually two- $\mathbf{q}$ . This is inferred from the sharp knee in velocity for the  $c_{66}$  shear mode seen at 18 T, which can only couple to a CDW having components of propagation and displacement in both directions in the  $\mathbf{a} - \mathbf{b}$  plane. Our present *zero-field* results show that o-II exhibits a dominant CDW modulation vector different in magnitude and direction from that inferred from the NMR data but similar to that seen by diffraction at other dopings [1–3]. Assuming that the CDW is responsible for the Fermi surface reconstruction, the weak doping dependence of QO frequencies [14,15] also suggests that o-II develops CDW order similar to adjacent dopings. We are ineluctably driven to seek a new explanation for the NMR data consistent with these other measurements.

First, we note that the onset fields and temperatures given by NMR [4] and ultrasonics [18] differ from those given by x-ray diffraction [1–3]. As is discussed extensively elsewhere [1,2], NMR (and ultrasonics) are relatively low-frequency techniques, and it seems likely that quasistatic CDWs become visible to x rays before they become slow enough to have strong effects on NMR and ultrasonics. It is possible that the anomaly seen at 18 T with ultrasound [18] is the locking of the CDW order that we observe into  $\delta \rightarrow 1/3$ , accompanied by a freezing of the CDW.

A one- $\mathbf{q}$  *incommensurate* sinusoidal charge density wave along  $\mathbf{b}$  would give a bimodal distribution of NMR frequencies at a given Cu site, with van Hove peaks at the extremal values, while a one- $\mathbf{q}$  *commensurate*  $\delta = 1/3$  modulation can give two peaks of unequal weight. If barely resolved, either could mimic a simple splitting. In contrast,

a two- $\mathbf{q}$  incommensurate pattern, with the two components identical, would give a single central van Hove peak. However, a two- $\mathbf{q}$  pattern with one component having rather weaker effects than the other could again give a bimodal distribution, similar to that observed for the Cu2F sites. A question for any modulation along  $\mathbf{b}$  is to explain the much smaller effects of the CDW on the Cu2E sites. This could arise if the CDW caused larger displacements of the atoms (e.g., oxygen O3) near the Cu2F sites, as is allowed by symmetry in the full o-II unit cell. Alternatively, the stronger effects of the CDW at the Cu2F sites may be related to their proximity to the conducting charge reservoirs represented by the well-ordered Cu-O chains. We put forward this model in an attempt to reconcile apparently conflicting results from different methods. At the present, diffraction data cannot be taken at sufficiently high steady magnetic fields to provide structural data to complement the spectroscopic data from NMR, the thermodynamic data from ultrasonics, and the Fermi surface data from QO.

In summary, we have detected CDW ordering in ortho-II YBCO in a zero magnetic field. The major component of the ordering has been found to have  $\mathbf{q}_{\text{CDW}}$  with an in-plane component along the  $\mathbf{b}$  (chain) direction. This contrasts with nearby higher dopings having less perfect Cu-O chain order, where two- $\mathbf{q}$  structures are observed in which the two modulations have similar amplitudes. The incommensurability and  $T$  dependence of the CDW order are very similar to those previously reported in o-VIII and III YBCO, but there is a clear trend to a larger  $q_{\text{CDW}}$  at lower doping; this suggests a band-structure influence on  $q_{\text{CDW}}$ . Our observation in o-II YBCO of a dominant charge modulation along  $\mathbf{b}$  strongly suggests that a simple combined spin and charge stripe picture may not be appropriate since the incipient spin correlations have a wave vector along  $\mathbf{a}$ . The independent values of the spin and charge correlation  $\mathbf{q}$  vectors over a range of dopings indicate that these have different origins in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub>. For YBCO o-II, the dominant modulation direction in a zero field and its incommensurability are completely different from those inferred from high-field NMR data. We propose an alternative explanation of these data.

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*Note added in proof.*—A recent preprint [27] reports a soft x-ray study of a similar o-II YBCO sample. The authors find a modulation along  $\mathbf{b}$ , together with a weaker modulation along  $\mathbf{a}$ . A weak modulation along  $\mathbf{a}$  has been

revealed by further hard x-ray experiments by ourselves at the more intense beamline P07 on PETRA-III at DESY. The value of  $\delta_1$  given by our measurements is in Table I and Fig. 4.

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