Supporting Information: Ideal charge density wave order in the high-field state of superconducting YBCO

H. Jang[a,1], W.-S. Lee[b,1], H. Nojiri[c], S. Matsuzawa[c], H. Yasumura[c], L. Nie[d], A. V. Maharaj[d], S. Gerber[e], Y.-J. Liu[a], A. Mehta[a], D. A. Bonn[f,g], R. Liang[f,g], W. N. Hardy[f,g], C. A. Burns[a,h], Z. Islam[i], S. Song[j], J. Hastings[j], T. P. Devereaux[b], Z.-X. Shen[b,d], S. A. Kivelson[d], C.-C. Kao[k], D. Zhu[j,2], and J.-S. Lee[a,1,2]

[a]Stanford Synchrotron Radiation Lightsource, SLAC National Accelerator Laboratory, Menlo Park, CA 94025; [b]Stanford Institute for Materials and Energy Science, SLAC National Accelerator Laboratory and Stanford University, Menlo Park, CA 94025; [c]Institute for Materials Research, Tohoku University, Katahira 2-1-1, Sendai, 980-8577, Japan; [d]Geballe Laboratory for Advanced Materials, Departments of Physics and Applied Physics, Stanford University, Stanford, CA 94305; [e]SwissFEL, Paul Scherrer Institut, 5232 Villigen PSI, Switzerland; [f]Department of Physics & Astronomy, University of British Columbia, Vancouver, British Columbia, Canada V6T 1Z1 ; [g]Canadian Institute for Advanced Research, Toronto, Ontario, Canada M5G 1Z8; [h]Department of Physics, Western Michigan University, Kalamazoo, MI, 49008; [i]The Advanced Photon Source, Argonne National Laboratory, Argonne, IL 60439; [j]Linac Coherent Light Source, SLAC National Accelerator Laboratory, Menlo Park, CA 94025; [k]SLAC National Accelerator Laboratory, Menlo Park, CA 94025

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Our proposed interpretation of the x-ray results involves two basic assumptions: 1) In the absence of disorder, the intrinsic electronic correlations favor the formation of a unidirectional, incommensurate CDW, and 2) there are large scale inhomogeneities in the sample, which we treat schematically as regions of "More" and "Less" disorder. In this Supporting Information, we discuss how such assumptions can lead naturally to a sharp crossover in the directionality and correlation length of the CDW as the magnetic field is increased. Figure 5 in the main text illustrates the basic physics - in the $\sigma - \Lambda$ plane where σ characterizes the strength of disorder and Λ is the strength of the CDW (an increasing function of increasing magnetic field), there is a sharp crossover from short ranged bidirectional CDW correlations to longer ranged, strongly unidirectional CDW correlations. In a tetragonal cuprate, this would be associated with a thermodynamic phase transition to a nematic state, a form of "vestigial" order. We assume that at low fields both More and Less disordered regions are in the isotropic phase, while at low temperatures above a critical field strength the Less disordered regions enter the nematic phase, thus exhibiting strong unidirectional character.

A classical effective field theory

While many of the major points that underlie our proposal follow on rather general grounds from the statistical mechanics of disordered systems, the most straightforward way to illustrate them is by turning to the solution of the effective model of an incommensurate CDW introduced in Ref. [S1]. We consider a classical effective field theory with two complex fields, $\psi_x(\vec{r})$ and $\psi_y(\vec{r})$, representing the slowly varying amplitude of a CDW at wave vectors $\vec{Q}_x = q\hat{x}$ and $\vec{Q}_y = q\hat{y}$, respectively. A biquadratic coupling of the form $2\Delta|\psi_x|^2|\psi_y|^2$ appears in the effective action, where we take $\Delta > 0$ which favors unidirectional (stripe) over bidirectional (checkerboard) order. This model can be solved in the self-consistent Gaussian approximation using the replica trick to treat the disorder. The results are controlled in a formal $N \to \infty$ limit in which ψ_x is an $O(N)$ vector; it has been shown to agree qualitatively with results of Monte-Carlo simulations on the same model for the physical ($N = 2$) case in Ref. [S2].

As we are only interested in qualitative results, we will simplify the problem at the expense of neglecting several material specific features of YBCO: We consider a model with one plane per unit cell and assume an interlayer coupling, $V_z > 0$, that favors in-phase interplane ordering. Moreover, we have not included the in-plane anisotropy of the CDW stiffness constants, *i.e.* in the notation of Ref. [S1] we have taken $\kappa_\parallel = \kappa_\perp = 1$. (The final equality amounts to a particular choice of units of in-plane length, $b = 1$.)

For this simplified model, (for in-plane momenta $k_x^2 + k_y^2 \ll q^2$) the CDW structure factor can be expressed as

$$\begin{aligned} S(\vec{k} + \vec{Q}_x) &= TG(\vec{k}, \mu + \mathcal{N}) + \sigma^2|G(\vec{k}, \mu + \mathcal{N})|^2 \\ S(\vec{k} + \vec{Q}_y) &= TG(\vec{k}, \mu - \mathcal{N}) + \sigma^2|G(\vec{k}, \mu - \mathcal{N})|^2 \end{aligned} \quad \text{[S1]}$$

where

$$G^{-1}(\vec{k}, \mu) = \mu + k_x^2 + k_y^2 + V_z[1 - \cos(k_z c)]. \quad \text{[S2]}$$

G can be recognized as the simple Ornstein Zernike form of the order parameter correlations in a generic system in the disordered phase proximate to a critical point. k_z is the out-of-plane dispersion and c is the c-axis lattice parameter. The only subtlety is that the effective chemical potential (μ) and the "nematic order parameter" ($\mathcal{N}$), are determined from the self-consistency equations,

$$\Lambda(T, H) = \int \frac{d\vec{k}}{(2\pi)^3}[S(\vec{k} + \vec{Q}_x) + S(\vec{k} + \vec{Q}_y)] \quad \text{[S3]}$$

where Λ is the mean squared amplitude of the CDW order parameter (assumed to be an otherwise known function of T and H), and

$$\mathcal{N} = \mathcal{N}_0 + 2\Delta \int \frac{d\vec{k}}{(2\pi)^3}[S(\vec{k} + \vec{Q}_y) - S(\vec{k} + \vec{Q}_x)] \quad \text{[S4]}$$

where $\mathcal{N}_0$ is an intrinsic nematicity (due to the orthorhombicity of the crystal). Again, for simplicity, we will henceforth report results in the limit $\mathcal{N}_0 \to 0^+$. In this case, $\mathcal{N} = 0$ is always a possible solution of the self-consistency equations. However, if the CDW ordering tendency is sufficiently strong (*i.e.* for $\Lambda(T, H) > \Lambda_c$ where the critical value Λ_c is itself a function of

[1]These authors (H.J., W.-S.L., and J.-S.L.) contributed equally to this work.

[2]To whom correspondence should be addressed. E-mail: dlzhu@slac.stanford.edu (D.Z.) or jslee@slac.stanford.edu (J.-S.L.)

T and σ), a second solution with $\mathcal{N} > 0$ is preferred; this is the nematic phase which spontaneously breaks the point-group symmetry. Consistent with general theorems, so long as $\sigma > 0$, the CDW order inferred from these equations always has a finite correlation length, *i.e. there is never long-range CDW order* in the presence of even weak quenched randomness.

This set of equations was analyzed under various circumstances in Ref. [S1]. Generally, as the system is tuned from the isotropic to the nematic phase in any fashion (for instance, by increasing Λ) several changes in the nature of the correlations onset very rapidly at the transition point: The in-plane correlation length, ξ_{ab}, grows substantially as does the inter-plane correlation length, ξ_c, so long as V_z is not too small. The degree of directionality, $S(\vec{Q}_y)/S(\vec{Q}_x)$, which is 1 in the isotropic phase, becomes rapidly much larger than 1 in the nematic phase. Simultaneously, the peak intensity, $S(\vec{Q}_y)$, shows the most dramatic increase of all.

As an illustrative example, we consider the simplest case of $T = 0$ and vanishingly small interplane [S3] coupling, $V_z \to 0$. In this limit, the various integrals can be evaluated analytically. The self-consistency equations (which one is to solve for μ and $\mathcal{N}$) become

$$\begin{aligned}\Lambda(0,H) &= \left(\frac{\sigma^2}{2\pi}\right)\left[\frac{\mu}{\mu^2-\mathcal{N}^2}\right]\\ \mathcal{N} &= 2\Delta\left(\frac{\sigma^2}{2\pi}\right)\left[\frac{\mathcal{N}}{\mu^2-\mathcal{N}^2}\right]\end{aligned} \qquad \text{[S5]}$$

from which it follows that the critical value of Λ for the occurrence of the nematic phase is

$$\Lambda_c = \frac{\sigma}{2\sqrt{\pi\Delta}}\ . \qquad \text{[S6]}$$

For $\Lambda < \Lambda_c$, $\mu = \sigma^2/2\pi\Lambda$ and the in-plane correlation lengths and intensities of the peaks at $\vec{Q}_x$ and $\vec{Q}_y$ are

$$\begin{aligned}\xi_{ab}(\vec{Q}_x) = \xi_{ab}(\vec{Q}_y) &= \frac{1}{\sqrt{\mu}} \quad \text{and}\\ S(\vec{Q}_x) = S(\vec{Q}_y) &= 2\pi\Lambda|\xi_{ab}(\vec{Q}_y)|^4\ .\end{aligned}$$

For $\Lambda > \Lambda_c$, $\mu = 2\Delta\Lambda$, $\mathcal{N} = 2\Delta\sqrt{[\Lambda^2-\Lambda_c^2]}$, and

$$\xi_{ab}(\vec{Q}_x) = \frac{1}{\sqrt{\mu+\mathcal{N}}}\ , \quad \xi_{ab}(\vec{Q}_y) = \frac{1}{\sqrt{\mu-\mathcal{N}}}\ , \qquad \text{[S7]}$$

and

$$\frac{S(\vec{Q}_y)}{S(\vec{Q}_x)} = \left[\frac{\xi_{ab}(\vec{Q}_y)}{\xi_{ab}(\vec{Q}_x)}\right]^4\ . \qquad \text{[S8]}$$

To address the growth of 3D correlations in the nematic phase, it is obviously necessary to include explicitly the effects of non-zero V_z. While the self consistency equations are still analytically tractable, the solutions are sufficiently complicated that we only evaluate them numerically. The inter-plane correlation length can be computed as

$$\xi_c = (c/2)\left[\operatorname{arcsinh}\left(1/\xi_{ab}\sqrt{2V_z}\right)\right]^{-1}. \qquad \text{[S9]}$$

Clearly, there is strong tendency to increased 3D order when the in-plane correlations become sufficiently long.

In Fig. S1, we show the evolution of these quantities computed numerically from the full self-consistency equations with $V_z = 0.1$ as a function of Λ for two different values of σ, one representative of the More and one of the Less disordered regimes, under the assumption $\sigma_{\text{More}}/\sigma_{\text{Less}} = 1.5$. To make contact with experiment, one should imagine that the field dependence of Λ determined by competition with superconductivity, $\Lambda = \Lambda_0[1-|\phi|^2]$, where the amplitude of the superconducting order ϕ is (presumably) a decreasing function of increasing H. There is clearly a sharp increase in the in-plane correlation lengths and peak intensities at the nematic phase transition.

Subtleties and ambiguities: One implication of "random-field-type" disorder is slow dynamics which inevitably result in the system falling out of equilibrium upon cooling before the point of the transition is reached. A rather subtle experimental protocol is necessary to establish the existence of a broken symmetry phase experimentally [S4]. However, YBCO is weakly orthorhombic, which in the current context means that there is effectively always a small symmetry breaking field; this rounds the transition to the nematic state, but at the same time reduces the tendency to the formation of a metastable nematic domain structure. (From a complementary perspective, it is possible to look for a growing thermodynamic correlation length by detecting [S5] the telltale hysteresis and noise characteristic of the random-field-Ising model in this limit [S6].)

One might still worry that the time-scales involved in a pulsed field experiment could exacerbate non-equilibrium effects. However, since the slow dynamics are associated with reorientation of growing domains [S7, S8], the characteristic relaxation rates depend exponentially on domain size. Therefore, once the experimental time scales are large compared to microscopic time scales (as they are in our experiment), an enormous increase in the time scale only permits one to probe the equilibrium physics of slightly larger domains.

Finally, it is tempting to try to estimate the volume fractions of the More and Less disordered regions from the relative strengths of their contributions to the x-ray scattering intensity, $I(\vec{k})$. However, making connection between the calculated and measured quantities carries with additional uncertainties. $I(\vec{k})$ is generally dominated by scattering of the atomic cores, and so is a measure of the atomic displacements. A periodic pattern of atomic displacements proportional to the CDW order parameter, ψ, is generic. However, while positions and widths of the observable peaks in $I(\vec{k})$ reflect the pattern of translation symmetry breaking and the CDW correlation length, the variation of intensity from one Brillouin zone to the other encodes the precise pattern of induced lattice distortion. Crudely,

$$I(\vec{k}) = I(\vec{K}+\vec{Q}) \approx I_0(\vec{K})S(\vec{Q}) \qquad \text{[S10]}$$

where $\vec{K}$ is a reciprocal lattice vector, and $I_0(K)$ is a complicated structure factor.

If the scattering intensities were measured in sufficiently many different Brillouin zones, one could work backwards to the actual lattice displacements induced by the CDW. This has been undertaken for the low field diffraction data in Ref. [S9]. Because of the constraints of the high field experiment, our data is limited to little over one Brillouin zone, so such a refinement is not possible. Even in the one Brillouin zone we study, we have not directly measured the peak width in all directions — our estimates of the correlation volume involve the implicit assumption that the correlation length is not much different in the a and b directions. For all these reasons, there is considerable unavoidable uncertainty in any estimate we might make of the relative volume fractions of the two regions. These worries aside, in the ortho-VIII sample at 25 T [S10], we find the integrated intensity in the 2D peaks is roughly 5 times that in the 3D peak suggesting that something between 10% and 20% of the sample is Less disordered.

S1 Nie L, Tarjus G, Kivelson SA (2014) Quenched disorder and vestigial nematicity in the pseudogap regime of the cuprates. *PNAS* 111(22):7980–7985.

S2 Nie L, Sierens LEH, Melko RG, Sachdev S, Kivelson SA (2015) Fluctuating orders and quenched randomness in the cuprates. *Phys. Rev. B* 92(17):174505.

S3 (Note) Strictly speaking, the nematic phase we find for $V_z = 0$ is an artifact of the large N approximation; random field disorder precludes even discrete symmetry breaking in 2D. However, as shown in ref. [1], for small but non-zero V_z, the nematic phase is robustly recovered, even when V_z is small enough that it has little effect on the quantities considered here.

S4 Birgeneau RJ, Shapira Y, Shirane G, Cowley RA, Yoshizawa H (1986) Random fields and phase transitions. *Physica B+C* 137(1):83–95.

S5 Bonetti JA, Caplan DS, Van Harlingen DJ, Weissman MB (2004) Electronic Transport in Underdoped $YBa_2Cu_2O_{7-\delta}$ Nanowires: Evidence for Fluctuating Domain Structures. *Phys. Rev. Lett.* 93(8):087002.

S6 Carlson EW, Dahmen KA, Fradkin E, Kivelson SA (2006) Hysteresis and Noise from Electronic Nematicity in High-Temperature Superconductors. *Phys. Rev. Lett.* 96(9):097003.

S7 Bruinsma R, Aeppli G (1984) Interface Motion and Nonequilibrium Properties of the Random-Field Ising Model. *Phys. Rev. Lett.* 52(17):1547–1550.

S8 Huse DA, Fisher DS (1987) Dynamics of droplet fluctuations in pure and random Ising systems. *Phys. Rev. B* 35(13):6841–6846.

S9 Forgan EM et al. (2015) The microscopic structure of charge density waves in underdoped $YBa_2Cu_3O_{6.54}$ revealed by X-ray diffraction. *Nat Commun* 6:10064.

S10 Gerber S et al. (2015) Three-dimensional charge density wave order in $YBa_2Cu_3O_{6.67}$ at high magnetic fields. *Science* 350(6263):949–952.

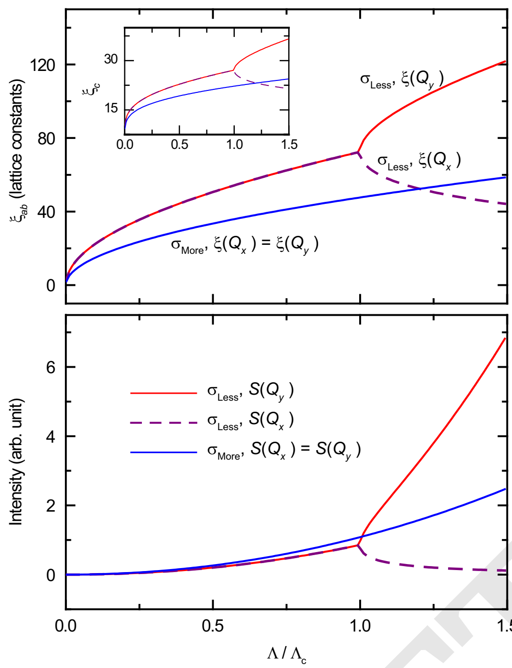

Fig. S1. Correlation lengths (upper panel) and peak intensities (lower panel) associated with the x and y directed components of the CDW order parameter as a function of increasing mean squared magnitude of the local CDW order parameter for the "Less" and "More" disordered regions. We have taken $\sigma_{\mathrm{More}} = 1.5\sigma_{\mathrm{Less}}$, $V_z = 0.1$, and for graphical clarity have rescaled S for the More disordered regions by a factor of 3. The main panel a shows the in-plane correlation lengths, while the inset shows the correlation length with the z-direction (*e.g.*, c-axis). Here, Λ_c is the critical value of the CDW magnitude in the Less disordered regions, and for simplicity we show results for $T = 0$. $\xi_{ab}(\boldsymbol{Q})$ and $\xi_c(\boldsymbol{Q})$ are the in-plane and out-of-plane correlation lengths corresponding to ordering vector $\boldsymbol{Q}$, respectively.

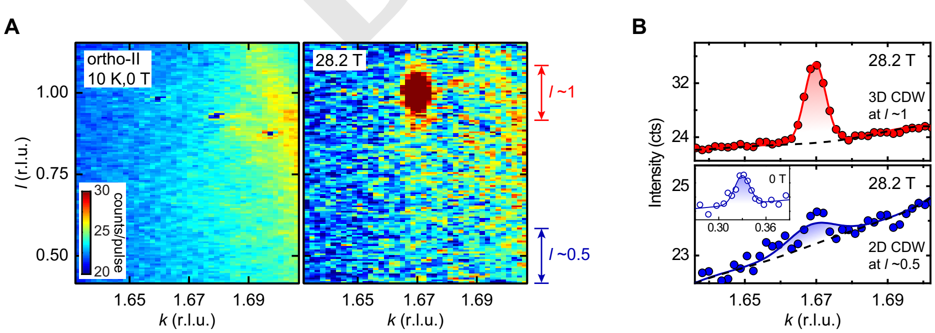

Fig. S2. (*A*) Diffraction intensity of ortho-II in the kl-plane near (0, 2-q, 1) at zero magnetic field (left panel) and at high magnetic field (right panel). Note that the feature around $k \sim 1.7$ and $l \sim 0.8$ is a condensation of water molecules results in the formation of an "ice ring" in the vicinity of the CDW signal [S10]. (*B*) Projected intensities from the diffraction pattern at $\mu_0 H$ = 28.2 T near $l \sim 1$ (upper panel) and $l \sim 0.5$ (lower panel). It indicates the coexistence of 3D and quasi-2D CDW orders in YBCO ortho-II under the high magnetic field. Inset (RSXS data) shows the quasi-2D CDW orders in YBCO ortho-II at $\mu_0 H$ = 0 T and $T \sim 20$ K. Colored solid lines are Gaussian fits to the high-field data with polynomial backgrounds at zero-field (black dashed line).

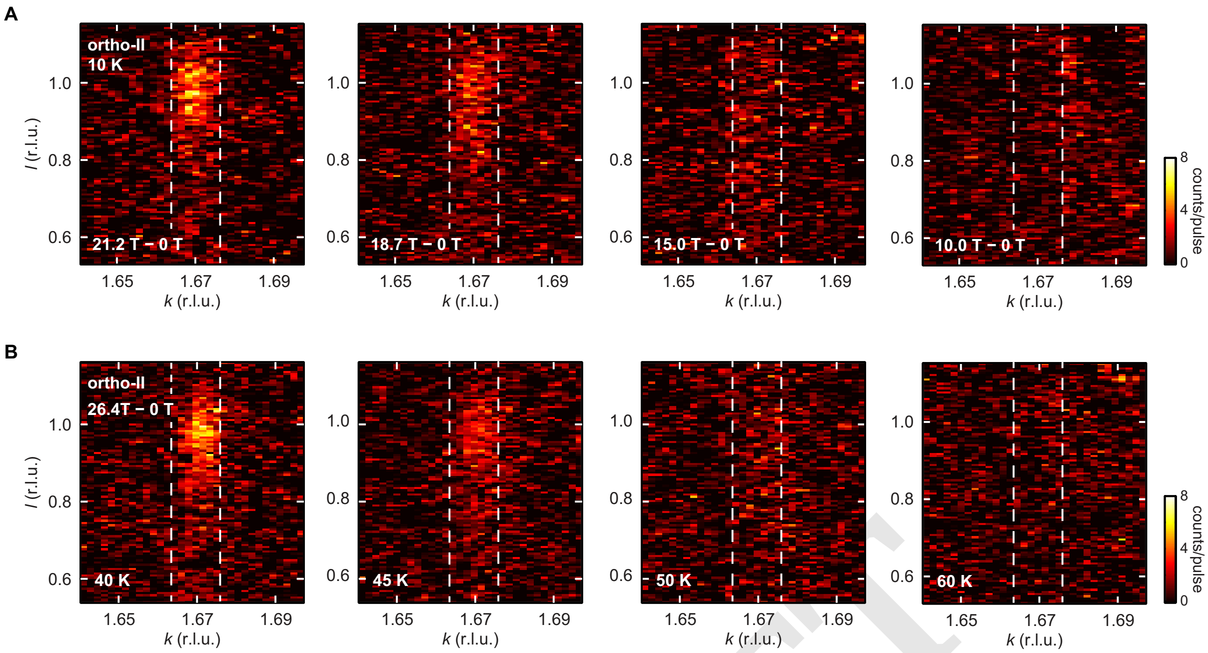

Fig. S3. (*A*) Magnetic field dependencies. (*B*) Temperature dependencies. Dashed lines indicate the windows used to deduce the projected peak profiles of Figs. 2A and 2D in the main text.

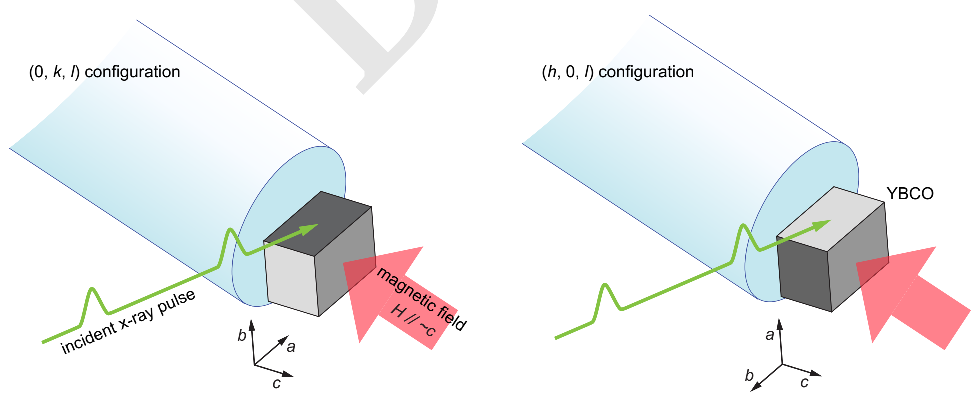

Fig. S4. Crystallographic orientations of YBCO crystals used for the measurement of the (0, *k*, *l*) reflection (left) and (*h*, 0, *l*) reflection (right). The green line represents the incident x-ray free electron pulses. The red arrow indicates the direction of the magnetic field.