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Quasiparticle scattering off defects and possible bound states in charge-ordered $YBa_2Cu_3O_y$

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We report the NMR observation of a skewed distribution of ^{17}O Knight shifts when a magnetic field quenches superconductivity and induces long-range charge-density-wave (CDW) order in $YBa_2Cu_3O_y$. This distribution is explained by an inhomogeneous pattern of the local density of states $N(E_F)$ arising from quasiparticle scattering off, yet unidentified, defects in the CDW state. We argue that the effect is most likely related to the formation of quasiparticle bound states, as is known to occur, under specific circumstances, in some metals and superconductors (but not in the CDW state in general, except for very few cases in 1D materials). These observations should provide insight into the microscopic nature of the CDW, especially regarding the reconstructed band structure and the sensitivity of disorder.

Recent experiments in high-T_c copper-oxides at moderate doping levels have led to a consensus regarding the presence of static, but essentially short-range, spatial modulations of the charge density that appear far above the critical temperature T_c and compete with superconductivity below T_c [1–12]. As a result, the debate has mainly focused on this "incipient" charge-density-wave (CDW) in zero magnetic field. Somewhat less attention has been paid to another CDW order that also coexists and competes with superconductivity but is present at lower temperatures, only in high magnetic fields, and has been detected only in $YBa_2Cu_3O_y$ (YBCO) so far, by nuclear magnetic resonance (NMR) [13, 14] and more recently by X-ray diffraction [15, 16]. This CDW develops long-range three-dimensional (3D) correlations above a critical field (also detected in sound velocity [17] and thermal conductivity measurements [18]). In principle, the long-range coherence of the high-field CDW makes it an ideal playground for investigating the nature of CDW order and its interplay with superconductivity [19]. However, since its observation requires high fields, experimental characterization of this phase has remained limited. This evidently prompts for further high field experiments in YBCO.

Here, we report that an anomalous spatial distribution of ^{17}O Knight shift values develops alongside with the high-field CDW in YBCO. We explain the effect by an enhancement of $N(E_F)$, the *local* density-of-states at the Fermi energy, around defects and we argue that this most likely due to the formation of quasiparticle bound states, as previously observed in the superconducting state of cuprates [20–27]. This unforeseen observation raises important questions on the electronic band structure and on the nature of defects in the CDW state.

^{17}O-enriched untwinned single crystals of $YBa_2Cu_3O_y$ were prepared with $y_{\mathrm{nominal}} = 6.47$ (ortho-II oxygen ordering, hereafter O-II), 6.56 (O-II), 6.68 (O-VIII) and 6.77 (O-III), following ref. [28]. Their high quality is attested by the very sharp NMR lines, their oxygen order is attested by the observation of inequivalent chain or planar sites [29] and their doping level is attested by the values of the superconducting, vortex melting and CDW transition fields/temperatures, which are all consistent with the literature [2, 14]. More information about samples and experimental methods can be found in supplemental material [30] and in refs. [2, 14, 29].

Fig. 1a shows a typical NMR spectrum in the field-induced CDW state of $YBa_2Cu_3O_{6.56}$, for O(2) and O(3) sites, which are those sites in CuO_2 planes lying in bonds oriented along the crystalline a and b axes, respectively [30]. Each site comprises five lines corresponding to the different nuclear transitions of the nuclear spin $^{17}I = 5/2$. Below ∼100 K, we observe that the separation between O(3E) lines (sites below empty chains) and O(3F) (below full chains) is much smaller than the linewidth and thus these sites are not distinguished.

For both O(2) and O(3), the low-frequency satellites (LF1 and LF2) present a clear asymmetric profile with a long tail toward high frequencies. The same asymmetry is also clear on the first high-frequency (HF1) satellite of O(3) as well as on the central line. Other lines are either split by charge order (O(2) HF1 and HF2) or they experience strong quadrupole broadening (O(2) and O(3) LF2), which in both cases makes the asymmetry, if any, less visible. Detailed analysis of these spectra [30] indicates that the asymmetry is actually present on each individ-

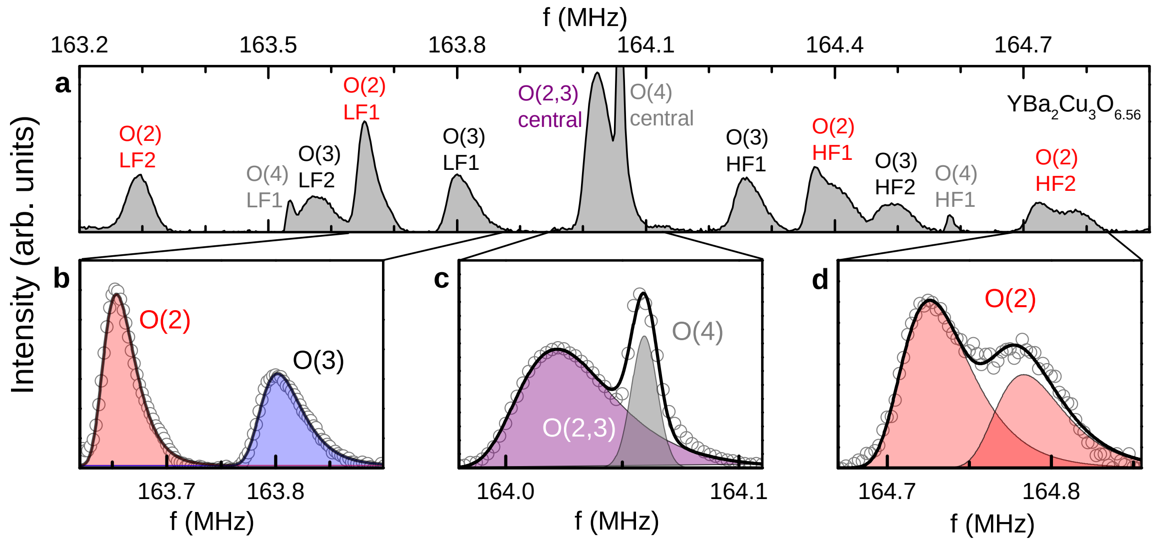

FIG. 1: (Color online). (a) ^{17}O NMR spectrum of $YBa_2Cu_3O_{6.56}$ at $T = 3$ K and $H \simeq 28.5$ T (27.4 T for the c-axis projection as the field is tilted off the c-axis [30]). HF1(2) are the first (second) high-frequency satellites and LF1(2) are the first (second) low-frequency satellites. For the apical O(4) site, HF2 and LF2 lines are out-of-scale. (b-d) Zoom on particular lines with fits using an extreme value distribution function (see text). The O(2) HF2 line is split by CDW order.

ual line and that it does not result from an unresolved line splitting on some of the lines. The asymmetric line broadening actually adds to both the splitting produced by long-range CDW order [13, 14] and the symmetric broadening due to short-range CDW order [2].

The main outcome of the analysis is that the asymmetry arises exclusively from a spatial distribution of local magnetic fields, not from a distribution of electric-field-gradients. The histogram of these local fields is directly given by the NMR lineshape, provided other CDW-induced effects are comparatively small (as is the case for LF1 lines, for example). Therefore, the triangle-shaped distribution points to an inhomogeneous state in which an overwhelming majority of sites experience small local fields whereas sites with large fields are relatively rare. This is the first central result of this work.

We quantify the asymmetry by fitting the lines with Gaussians having distinct right and left widths w_R and w_L ($w_R \geq w_L$). As shown in Fig. 2a,b, all of the asymmetry arises from the broadening of the right (high-frequency) part of the line, meaning that the distribution involves only enhanced values of the local field. Furthermore, both the field and the temperature dependence w_R (or equivalently of the asymmetry $A = (w_R - w_L)/(w_R + w_L)$) closely follow the variation of the ^{17}O line splitting that provided direct evidence of CDW order in high fields [14]. Therefore, the spatial distribution of local fields arises only in conjunction with long-range CDW order. This is the second central result of this work.

The asymmetric boadening at $T = 3$ K becomes field-independent above $\sim$25 T (Fig. 2c,d). At these fields, the bigaussian with $A \simeq 0.35$ is actually very close to an extreme value distribution (EVD) function, $f(x) = b\exp(-\exp(-z) - z + 1)$ with $z = (x - x_c)/w$, found in a variety of critical phenomena [31–36]. Quite remarkably, this asymmetry in high fields is found to be the same in four samples having not only different oxygen contents but mostly different levels of disorder, as indicated by the factor of 3 difference in their high temperature linewidth $w_{\rm HT}$ [30]. The identical asymmetry for all samples simply follows from the fact that both w_R and w_L are proportional to $w_{\rm HT}$ [30]. This strongly suggests that the asymmetric broadening is triggered by disorder. This is our third main observation.

That the anomalous distribution is tied to long-range CDW order disorder immediately suggests that it is unrelated to the vortex lattice or to magnetic order. This is confirmed by the following observations. First, vortex broadening typically decreases with increasing field, contrary to what we observe here. Second, the distribution is observed up to at least 45 T while the vortex lattice melts near $B_{\rm melting} \simeq 24$ T at $T = 3$ K [37]. Therefore, in most of the field and temperature range investigated, vortices fluctuate much faster than our typical spectral resolution of $\sim$10 kHz and thus cannot broaden the NMR spectrum. For the same reason, any electronic pattern associated with the vortex cores (such as Andreev bound states) cannot explain the results. Last, if due to magnetic order, the broadening should be preceded by an enhancement of the relaxation rates, which is not seen in either ^{63}Cu or ^{17}O data.

Having excluded broadening by vortices and magnetic order, it is then appropriate to speak in terms of inho-

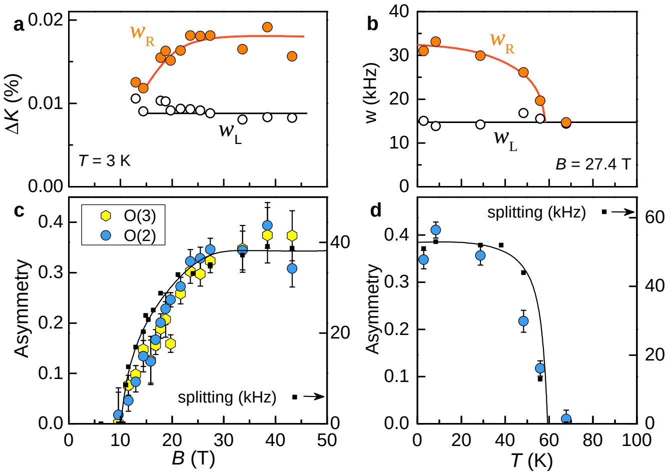

FIG. 2: (Color online). (a,b) Field and temperature dependence of w_R and w_L, the right (high-f) and left (low-f) width of the O(2) LF1 line in $YBa_2Cu_3O_{6.56}$. (c,d) Field and temperature dependence of the line asymmetry for O(2) and O(3) LF1 lines, calculated from data in (a) and (b), and compared to the quadrupole part of the O(2) HF2 line splitting $\Delta\nu_Q$ from ref. [14]. The field was tilted off the c-axis toward b in (a,c) and toward a in (b,d).

mogeneous Knight shift (K) of a paramagnetic metal. K is defined as the shift $K = (f - f_0)/f_0$ of the resonance frequency f with respect to a reference f_0, due to the local magnetic susceptibility. For $B \parallel \alpha$ where $\alpha = a, b, c$ represents the crystallographic axes of YBCO, the diagonal components of the Knight shift tensor K are related to the static, uniform, spin susceptibility $\chi^{\rm spin} = \chi^{\rm spin}(q = 0, \omega = 0)$ through:

$$K_{\alpha\alpha}(T) = K^{\rm spin}_{\alpha\alpha}(T) + K^{\rm orb}_{\alpha\alpha} = \frac{A^{\rm hf}_{\alpha\alpha}}{g_{\alpha\alpha}\mu_B}\chi^{\rm spin}_{\alpha\alpha} + K^{\rm orb}_{\alpha\alpha} \quad (1)$$

where $A^{\rm hf}$ is the hyperfine tensor and g the Landé factor. $K^{\rm orb}$ is mostly attributed to Van-Vleck paramagnetism.

The continuous aspect and the skewness of the lineshapes suggest that K is maximal at relatively few locations and that it decays over a typical distance much larger than a lattice step. There are not many examples of such Knight shift distributions. For instance, oxygen vacancies in the chains or impurities in the planes produce a staggered magnetization that leads to symmetric NMR broadening [20, 38, 39]. An asymmetric charge density distribution (that could result, for example, from the interference between the CDW and Friedel oscillations [40]) would produce an asymmetric distribution of K. However, this should also produce a measurable quadrupole effect, because charge variations in cuprates produce frequency shifts that are of the same order of magnitude in the quadrupole and Knight shift channels (~ 1 MHz/hole for ^{17}O in $B \sim 10$ - 30 T). Therefore the absence of any measurable quadrupole broadening tied to the skewed distribution of K [30] indicates that the latter is not related to a charge modulation. In particular, it cannot be a direct imprint of the CDW, which is known to produce a line splitting contributed about equally by bimodal distributions of K and of the quadrupole frequency [13, 14].

In metallic systems, the density-of-states at the Fermi level $N(E_F)$ can be *locally* enhanced without direct connection to a change in the charge density. Since $N(E_F)$ enters into $\chi^{\rm spin}$, K is locally enhanced (Eq. 1) and the probability distribution of K values (*i.e.* the NMR lineshape) becomes skewed. This occurs in the presence of Anderson localization [41, 42], that should be irrelevant here, or in the presence of various types of in-gap (bound) states.

The recent proposal of a pair-density-wave (PDW) state in the cuprates [43–46] provides a tantalizing justification of Andreev bound states at locations where the superconducting gap changes its sign. Zero-bias conductance peaks in scanning-tunnelin-microscopy (STM) have actually been interpreted as such evidence in $La_{1.88}Sr_{0.12}CuO_4$ [47] (see also Refs. [48, 49] for zero-bias conductance peaks in 2D chalcogenides). However, our anomalous K distribution persists up to at least 45 T (Fig. 2), far above $H_{c2} \simeq 24\pm$ 2 T in $YBa_2Cu_3O_{6.56}$ [50–52]. In order to explain our results, Andreev bound states would then have to be pinned by disorder and to survive in the metallic but non-superconducting state above H_{c2}. While it has been envisaged that a phase-disordered [53] or short-ranged fluctuating [44] PDW state could survive above H_{c2}, none of these situations is likely to produce static bound states. Even if Therefore, this scenario

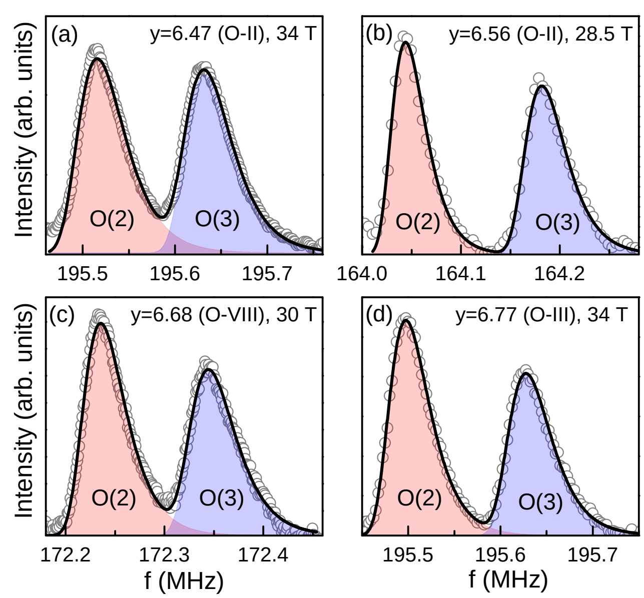

FIG. 3: (Color online). O(2) and O(3) first low frequency quadrupole satellites showing similar asymmetric profile for four different samples (see methods). Continuous lines are fits of each peak to an extreme value distribution (see text).

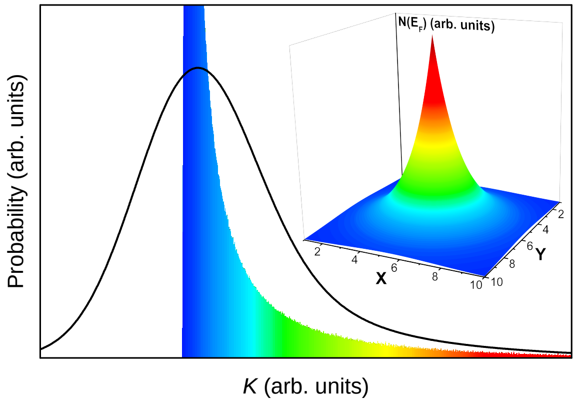

FIG. 4: (Color online). Probability distribution of the Knight shift $K \propto N(E_F)$ for the real space pattern of $N(E_F)$ shown in inset (exponential decay around a given position in the (x, y) plane). The black line is the histogram convoluted with a Gaussian distribution.

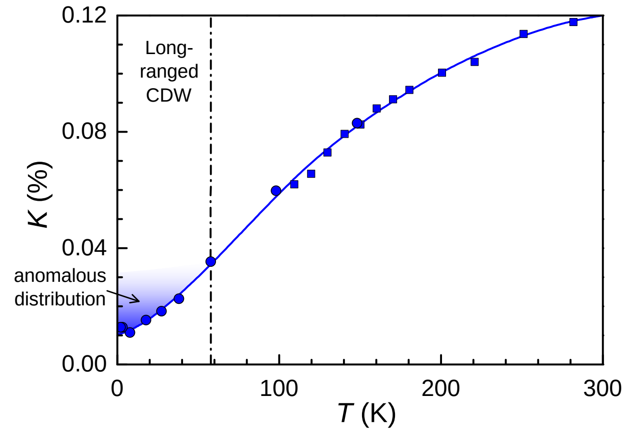

FIG. 5: (Color online). Knight shift K measured at 28.5 T (circles) and its asymmetric distribution in $B \gtrsim$ 20 T, schematically represented by the colored area ($B \parallel c$, $YBa_2Cu_3O_{6.56}$). K is defined as the most probable value of the distribution, *i.e.* at the position of maximum intensity on the line of O(2,3) sites. Squares are shift data measured at 12.0 T. Above 150 K, O(2) and O(3) central lines can be resolved so K is defined as the average shift of the two lines. The line is a guide to the eye.

seems unable to explain our data.

Defect-induced bound states have been observed in the chains of $YBa_2Cu_3O_{6.97}$ [54] and so one could speculate that we are seeing here their impact onto the planes, perhaps facilitated by the transverse coherence of the CDW above $H^{3D}_{\rm charge}$ ($\simeq$ 15 T near $p = 0.11$ doping [16]). This is however unlikely as no asymmetric ^{17}O broadening has been reported in $YBa_2Cu_3O_{6.97}$ (besides vortex broadening at low T) and furthermore the asymmetric broadening appears at $H^{2D}_{\rm charge} \simeq 10$ T well below $H^{3D}_{\rm charge}$.

Finally, there remains one explanation for which we find no counterarguments: defect-induced electronic bound states in CuO_2 planes, even tough these are not known to be a generic property of the CDW state. Such "Impurity resonances" around defects have been documented by scanning tunneling microscopy (STM) in the superconducting state of cuprates [22–26], or in their pseudogap state [27]. Furthermore, the asymmetric profile of our ^{17}O NMR lines is strikingly similar to that reported in Zn-doped $YBa_2Cu_3O_7$, including an asymmetry that is independent of the amount of disorder [30]. The broadening of the high-frequency side has been shown [20, 21] to directly reflect the real-space profile of the bound state, such as depicted in Fig. 4.

In general, observing how electronic states in solids react to a local symmetry breaking provides insight into their microscopic nature. Below we argue that, although it primarily raises many questions, our observation could provide an insightful window into charge ordering. Note that, since it is very likely that the effect we observe is produced by scattering off some disorder, many of the considerations below would hold even if an alternative explanation to quasiparticle bound states were to be found.

First, identifying the nature of the defects responsible for scattering is an important task. Chain-oxygen defects have been shown to constitute the main source of electronic scattering [55] and they are indeed ubiquitous in the chain layer of oxygen-ordered YBCO [29]. Therefore, it is likely that scattering is related to out-of-plane disorder rather than to in-plane impurities or vacancies. On the other hand, the separate onsets for asymmetric broadening ($H^{2D}_{\rm charge} \simeq 10$ T) and for c-axis coherence ($H^{3D}_{\rm charge} \simeq 15$ T [16, 17]) suggest that quasiparticles are not scattered directly off out-of-plane defects. This paradox may be resolved if the chains are involved only indirectly in the scattering, that is, via electronic perturbations created in the planes by chain defects. Possible candidates are: phase slips or amplitude defects of the CDW, Friedel oscillations [56], patches of uncondensed short-range CDW order [2, 15, 16] and patches of short-range spin-density-wave order (long-range CDW order triggers slow fluctuations [13] that could be pinned by disorder and promote a Kondo resonance [57]).

In-gap states, attributed to solitonic defects, have been reported in one-dimensional CDW materials [58–60]. If the defects here are phase slips, it is the possible that the incommensurability seen by X-ray scattering in high fields is due to the presence of domain walls (discommensurations) that separate domains of locally commensurate CDW order. This would then suggest that charge modulation are rooted in the doped-Mott insulator nature of the cuprates [61]. Also important is the elucidation of the role of defects in the chain layer. This could shed light on a hypothetical role of the chains in the high-field CDW [62] as well as on the nature of pinning of the short-range CDW [2, 63, 64].

Clarifying which aspect of the CDW is crucial in the formation of bound states should be informative on its microscopic nature. Since neither the pseudogap (that indeed persists in the high field CDW [14]) nor the substantial CDW modulations of the normal state (the correlation length ξ_{ab}^{CDW} reaches 20 lattice spacings) appear to be sufficient conditions for the effect observed here, it must be that either the large values of ξ_{ab}^{CDW} or the uniaxial nature of the field-induced CDW [13, 16] (or both) are pivotal. As already mentioned, the c-axis coherence does not seem to be connected to our observations. Another interesting question is whether a putative, but likely, d-wave symmetry of the intra-unit-cell form factor [65, 66] of the high-field CDW could play any role. Theoretically, bound states have been found, under certain conditions, in models of one-dimensional CDW [67, 68] and in the d-density-wave state [69–74], but they do not occur at E_F (note that they only need to have a finite weight at E_F to contribute to the Knight shift). Experimentally, impurity bound states have been observed in iron-based [75, 76] and heavy fermion [77] nodal superconductors as well as in graphene [78] and at the surface of topological insulators [79], so they have been argued to be a generic property of metals with Dirac-type electronic dispersion [80]. The presence of a Dirac cone in the band structure would then provide clues on the reconstructed Fermi-surface in high fields [81].

The results presented here call for theoretical investigations of the effects of disorder in two and three-dimensional CDW models for the cuprates. They should also stimulate further experimental work. In particular, a direct confirmation of the bound states, and possibly an identification of the defects, should come from STM experiments in high fields while some aspects of disorder pinning could be addressed by X-ray experiments [40].

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